Plastic mulch in agricultural systems: Degradation, fragmentation and transport with surface runoff

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Abstract

Agricultural plastics (e.g., greenhouses, tunnels, and mulch films) are indispensable in many parts of modern agriculture. They enhance crop yields by improving growing conditions and enabling year-round production. Mostly, they are made of resistant materials, such as polyethylene. Due to their persistence, plastic residues accumulate in soil, increasing agricultural soil pollution and negatively affecting soil and plant health. Additionally, once accumulated in soil, these residues can be transported to surrounding ecosystems (e.g., aquatic environments) *via*, for instance, surface runoff. One way to avoid the plastic residue accumulation in soils is to use soil-biodegradable mulch films. However, as the full biodegradation in soil takes years, soil-biodegradable residues can also be transported to surrounding ecosystems. Due to the many variables and effects that agricultural plastics can have on the environment, this thesis is divided into three main sections, aiming to elucidate main research gaps regarding (1) the degradation and (2) influencing factors on the accumulation of conventional agricultural plastics, as well as (3) the transport and soil effects of soil-biodegradable alternatives.

Understanding the degradation and accumulation factors of agricultural plastics is essential to elucidate the environmental abundance of plastic residues and to develop effective mitigation strategies to avoid and control soil pollution. Hence, the first main section of the thesis is based on a controlled laboratory experiment, where photodegradation *via* ultraviolet radiation and mechanical abrasion of different types of low-density polyethylene agricultural plastics over different time periods was analysed. The analysis underlined that photodegradation and the removal of microplastic surface particles through mechanical abrasion already occurred after short ultraviolet radiation exposure. Abrasion has been identified as an important modulator in photodegradation, as it removes photodegraded surface particles, thereby exposing undegraded film surfaces to ultraviolet radiation.

The second main section of the thesis focuses on analysing field samples to understand the influence of different regional climates (Germany vs. Spain) and management practices on the concentrations of macroplastics (> 1 cm) and microplastics (1–5 mm) in agricultural soil systems. Regional differences were evident, with Spanish fields generally presenting higher plastic concentrations than German fields, likely due to differences in climate and/or management practices (i.e., compost and plastic characteristics). The results of this study highlight that plastic contamination in agricultural soils cannot be attributed only to the use of mulch films, but rather to a combination of factors (i.e., intensity of plastic use, persistence of different plastic products, soil management, as well as other environmental factors like site-specific ultraviolet radiation). Also, the diversity of methodological approaches across studies often hinders comparability. More standardised methodologies are necessary to increase comparability and isolate and identify influences of different factors (e.g., input source, management, or climate) in a holistic way.

Finally, the third section of the thesis focuses on field rainfall experiments to elucidate the effects of soil-biodegradable residues on surface runoff, soil erosion, and residue loss during heavy rainfall events. In this experiment, a significant increase in the runoff coefficient was caused by the soil-biodegradable residues, followed by a small transport of small residues, even though the sediment delivery didn't present a significant change between treatments. However, it is expected that surface runoff will diminish as larger residues continue to biodegrade, while smaller residues may be transported during erosion events, as long as they remain in the topsoil.

In summary, the knowledge gained from this PhD thesis provides a better understanding of the mechanisms and factors influencing plastic concentration in agricultural soil, as well as the impact of soil-biodegradable mulch films on residue transport and soil erosion processes. It has been demonstrated that the degradation and fragmentation of agricultural films are complex processes influenced by various factors. Additionally, it has been shown that climatic influences and different agricultural management practices affect the accumulation and distribution of plastic residues in agricultural soil. Furthermore, it has been demonstrated that the use of soil-biodegradable mulch films affects soil erosion and hydrological processes, and their residues are transported to surrounding ecosystems. This shows that the use of soil-biodegradable mulch films still presents concerns that must be considered, as it impacts not only the agricultural ecosystems but possibly other ecosystems as well (e.g., aquatic ecosystems). The insights of this thesis emphasise the importance of considering and understanding degradation processes, management practices, climate, and transport mechanisms, and their influence on soil erosion, when evaluating environmental impacts and mitigation policies.

Zusammenfassung

Agrarfolien (z.B. für Gewächshäuser, Folientunnel und Mulchfolien) sind in vielen Bereichen der modernen Landwirtschaft kaum noch wegzudenken. Sie steigern die Ernteerträge, indem sie die Wachstumsbedingungen verbessern und den Anbau von Nutzpflanzen mehr oder weniger ganzjährig ermöglichen. Die meisten Agrarfolien bestehen aus Polyethylen (PE). Aufgrund der Persistenz der verwendeten Polymere und der oft kaum möglichen vollständigen Beseitigung nach der Verwendung reichern sich Folienfragmente im Boden an. Das kann je nach Polymertyp, verwendeten Aditiven sowie Fragmentmenge und -größe zu erheblichen Bodenbelastungen führen und letztendlich die Boden- und Pflanzengesundheit nachteilig beeinflussen. Darüber hinaus können die im Boden akkumulierten Plastikrückstände durch Wind und Oberflächenabfluss in umliegende Ökosysteme (z.B. Gewässer) eingetragen werden. Um eine nachteilige Akkumulation persistenter Kunststofffragmente im Boden zu vermeiden, werden zunehmend bioabbaubare Materialien verwendet, die nach der Nutzung in den Boden eingearbeitet werden. Da der avisierte vollständige Abbau im Boden jedoch Jahre dauert, können auch biologisch abbaubare Folienstofffragmente in umliegende Ökosysteme gelangen, in denen andere Abbaubedingungen herrschen.

Aufgrund der vielfältigen Einflussfaktoren und der Auswirkungen von Agrarfolien auf die Umwelt wurden in der vorliegenden Arbeit drei Aspekte näher beleuchtet, um zentrale Forschungslücken zu schließen: (1) die Prozesse der Degradierung und Fragmentierung durch UV-Strahlung und mechanische Einflüsse, (2) die Einflussfaktoren auf die Akkumulation konventioneller Agrarkunststoffe und (3) der Einfluss von Folienresten auf den Oberflächenabfluss und die Erosion. Das Verständnis der Abbau- und Akkumulationsfaktoren von Agrarfolien ist essenziell, um die Umweltbelastung durch Kunststoffrückstände zu bestimmen und darauf aufbauend wirksame Strategien zur Vermeidung der Bodenkontamination zu entwickeln.

Im ersten Hauptteil der Arbeit wurden unter kontrollierten Laborbedingungen Analysen zum photochemischen Abbau und zum mechanischen Abrieb verschiedener Arten von Polyethylen-Agrarfolien niedriger Dichte über unterschiedliche Zeiträume durchgeführt. Dabei wurde beobachtet, dass Photodegradation und die Entfernung von Mikroplastikpartikeln durch mechanischen Abrieb bereits nach kurzer UV-Bestrahlung stattfinden. Die mechanische Belastung erwies sich dabei als entscheidender Faktor beim photochemischen Abbau, da diese zu einem Abrieb der photochemisch abgebauten Oberflächenpartikel führt und damit bisher nicht der UV-Strahlung ausgesetzte Polymerschichten der UV-Strahlung aussetzt.

Der zweite Hauptteil der Arbeit konzentriert sich auf die Analyse von Feldproben, um den Einfluss unterschiedlicher regionaler Klimata (Deutschland vs. Spanien) und Bewirtschaftungsmethoden auf die Konzentrationen von Makroplastik (> 1 cm) und Mikroplastik (1–5 mm) in landwirtschaftlichen Böden zu untersuchen. Regionale Unterschiede waren deutlich erkennbar: Die untersuchten Felder in Spanien wiesen im Allgemeinen höhere

Plastikkonzentrationen auf als die in Deutschland, was wahrscheinlich auf Unterschiede im Klima und/oder in den Bewirtschaftungsmethoden (z.B. Kompostnutzung, Frequenz der Foliennutzung, Verwendung unterschiedlich dicker Mulchfolien) zurückzuführen ist. Die Ergebnisse der Analysen verdeutlichen, dass die Plastikbelastung landwirtschaftlicher Böden nicht allein auf die Verwendung von Mulchfolie zurückgeführt werden kann, sondern vielmehr auf ein Zusammenspiel verschiedener Faktoren darstellt (z.B. Intensität der Plastiknutzung, Persistenz verschiedener Plastikprodukte, Bodenbewirtschaftung sowie andere Umweltfaktoren wie standortspezifische UV-Strahlung). Für eine Überblicksanalyse erschwert die Vielfalt methodischer Ansätze in verschiedenen Studien die Vergleichbarkeit.

Der dritte Hauptteil der Arbeit fokussiert sich auf den Einfluss von degradierbaren Folienfragmenten auf die Bildung von Oberflächenabfluss und Bodenerosion sowie auf den damit einhergehenden Abtrag von degradierbaren Mulchfolienfragmenten, die nur hinsichtlich des Abbaus für Böden als degradierbar zertifiziert sind. Bei Beregnungsversuchen, die Starkregen simulieren, verstärken die in den Boden eingearbeiteten Mulchfolienfragmente den Oberflächenabfluss, es zeigt sich jedoch kein signifikanter Effekt auf die Erosion. Da die Mulchfolienfragmente beim Versuch noch relativ groß waren (Zentimeterbereich), wurden nur geringe Mengen mit dem Oberflächenabfluss ausgetragen. Es ist davon auszugehen, dass der abflussverstärkende Effekt bei weiterer Degradation der Folienfragmente abnimmt und diese dann in Form von kleineren Fragmenten leichter mit dem Oberflächenabfluss bzw. der Erosion ausgetragen werden können.

Zusammenfassend liefert die vorliegende Dissertation Erkenntnisse zu einem besseren Verständnis der Mechanismen und Faktoren, die die Kunststoffkonzentration in landwirtschaftlichen Böden beeinflussen, sowie der Auswirkungen biologisch abbaubarer Mulchfolien auf den Transport von Pflanzenresten und die Bodenerosion. Es konnte gezeigt werden, dass der Abbau und die Fragmentierung landwirtschaftlicher Folien komplexe Prozesse sind, die von verschiedenen Faktoren beeinflusst werden. Darüber hinaus wurde nachgewiesen, dass klimatische Einflüsse und unterschiedliche landwirtschaftliche Bewirtschaftungsmethoden die Anreicherung und Verteilung von Kunststoffresten im Boden beeinflussen. Weiterhin wurde gezeigt, dass die Verwendung biologisch abbaubarer Mulchfolien die Bodenerosion und hydrologische Prozesse beeinflusst und deren Rückstände in umliegende Ökosysteme transportiert werden. Dies verdeutlicht, dass die Verwendung biologisch abbaubarer Mulchfolien weiterhin Bedenken aufwirft, die berücksichtigt werden müssen, da sie nicht nur landwirtschaftliche Ökosysteme, sondern möglicherweise auch andere Ökosysteme (z. B. aquatische Ökosysteme) beeinträchtigt. Die Erkenntnisse dieser Arbeit unterstreichen die Wichtigkeit, bei der Bewertung von Umweltauswirkungen und Minderungsmaßnahmen Degradationsprozesse, Bewirtschaftungspraktiken, Klima und Transportmechanismen sowie deren Einfluss auf die Bodenerosion zu berücksichtigen und zu verstehen.

Contents

General introduction 1

1.1 Plastics: the good and the bad side of the same coin

Due to their different properties and low cost, plastics are suitable for replacing a wide range of materials across many applications. Ceramic, glass, and metal are replaced by plastics in packaging, construction, the automotive industry, and agricultural, farming, and gardening sectors (Plastics Europe, 2023). With the mass production of plastic beginning during World War II (Callister & Rethwisch, 2018), the global production reached 413 × 10⁹ kg in 2023 (Plastics Europe, 2024b), where the most produced plastic type was polyethylene (low-density, linear low-density, medium-density, and high-density), accounting for 26.2% of total plastic production.

Despite their attractive properties and usability, plastic residues are now everywhere, from remote areas (Lavers & Bond, 2017) to groundwaters (Ren et al., 2021). Meijer et al. (2021) estimated the global annual emission of plastic into the ocean is between 0.8 to 2.7 × 10^9 kg per year. Meanwhile, the estimations of the global plastic emissions to the terrestrial environment made in 2016 ranged from 13 to 25 × 10⁹ kg per year, and it is expected that these estimates will double by 2025 (MacLeod et al., 2021). Even though the ranges and estimates can vary across studies, and are associated with large uncertainties, monitoring plastic residues across different ecosystems is crucial to determine their pollution status, concentrations, trends, and exposure to different organisms (Shim et al., 2017).

This thesis arises from the urgent need to increase our understanding of plastic pollution in agricultural soils. Before diving into the topic, some important terms need to be defined.

1.2 Definition: conceptual basis

1.2.1 Plastic

Plastic materials present structural rigidity and are, most of the time, composed of hydrogen and carbon (hydrocarbons) (Callister & Rethwisch, 2018). They can originate from natural sources, such as polylactic acid or synthetic ones, such as polyethylene (Callister & Rethwisch, 2018). Plastics can have a wide variety of properties, including rigidity, durability, flexibility, lightness, and versatility, among other good mechanical properties, and can also contain additives (Callister & Rethwisch, 2018).

Some of the desirable properties, such as elasticity, strength, thermal stability, optical properties (light transmission, colour, and opacity), and resistance to ultraviolet radiation (Hann et al., 2021; Steinmetz et al., 2016) are enhanced or achieved by adding additives, e.g., filler materials, stabilisers, antioxidants, plasticisers, and pigments (Callister & Rethwisch, 2018; Orzolek, 2017).

1.2.2 Classifications

Plastic can be classified based on its size (Ren et al., 2021; Zhang et al., 2021). Microplastics are plastics that typically fall below 5 mm in size (Ho et al., 2024). However, as the definition of microplastic has not yet been agreed upon, some authors have suggested that terms as nano-, meso-, and macroplastic should be used to define plastic particles as well (Hartmann et al., 2019). Due to the interdisciplinary nature of this thesis and the collaborative work of some chapters, a common size range for the entire thesis was not agreed upon. Therefore, the size ranges used are defined in each chapter.

Moreover, microplastics can be classified by origin. Primary microplastics are plastics produced on a micro scale for use, among other applications, in the cosmetics industry, pharmaceutical industry, and abrasives (R. Qi et al., 2020). Depending on the application, primary microplastics can vary in shape, size, and composition. Secondary microplastics, on the other hand, are microplastics resulting from the fragmentation of bigger plastics (R. Qi et al., 2020). Since secondary microplastics originate from the fragmentation of larger plastic materials, understanding degradation and fragmentation processes is necessary.

1.2.3 Bioplastics

Bioplastics are plastic materials with different properties and applications that are either biobased, biodegradable, or have both properties (European Bioplastics, 2022) (Figure 1.1). According to European Bioplastics (2022), biobased materials are derived, at least partly, from plants, such as corn, sugarcane, or cellulose, while biodegradable materials are converted by microorganisms into natural substances, such as water, carbon dioxide, and biomass, without the addition of additives. Biodegradability is linked to the chemical structure of the plastic material, and not to its origin (biobased or fossil-based). Therefore, biobased materials do not necessarily biodegrade, while some fossil-based materials can biodegrade (Figure 1.1) (European Bioplastics, 2022).

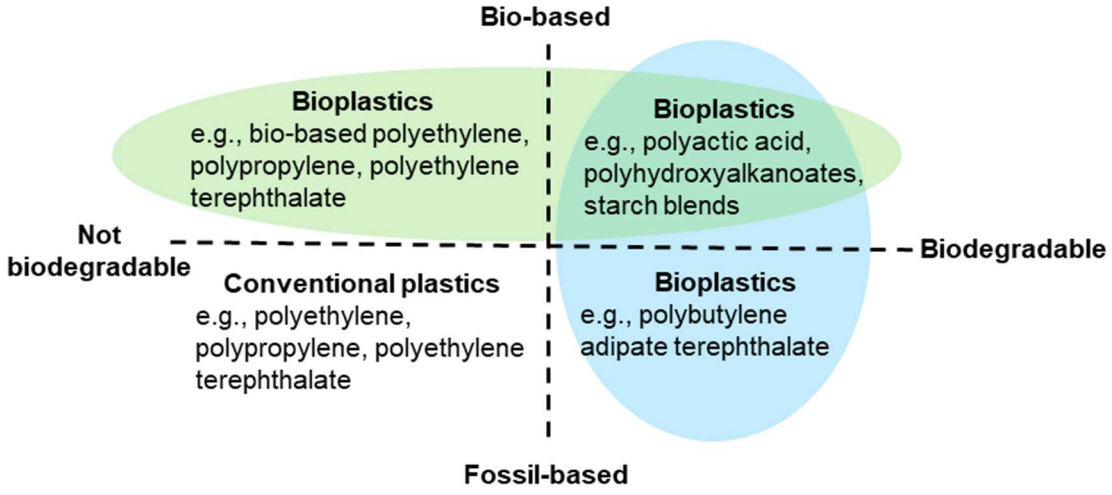

Figure 1.1: Diagram of bio-based, biodegradable, fossil-based plastic, and bioplastic. Adapted from European Bioplastics (2022).

1.3 Plastic in agricultural systems

Globally, agricultural plastic use has reached around 15.6 × 10^9 kg in 2021 (Plastics Europe, 2022). They can be used as mulching films, greenhouse covers, small tunnels, and other applications to enhance crop productivity (FAO, 2021). Agricultural plastics have become indispensable for intensive crop production, as they enable year-round production of fruits and vegetables, even under extreme weather conditions (Orzolek, 2017).

Plastic use in agriculture is referred to as plasticulture (Orzolek, 2017), and its widespread use started in the 1950s, with the use of low-density polyethylene as mulch film (Espí et al., 2006). The use of mulch films, for example, improves water use efficiency, regulates soil temperature, suppresses weeds, reduces the use of pesticides and fertilisers, and enables soil disinfestation with solar energy (soil solarisation), among other benefits (Agriculture Plastics Environment, 2021; Espí et al., 2006; Orzolek, 2017). Besides their benefits in crop production, agricultural plastics can also protect soil structures (Agriculture Plastics Environment, 2021), reducing soil compaction, as the soil will remain loose, friable, and well-aerated (Orzolek, 2017).

Despite its benefits, agricultural plastic waste has become a problem. Most of these plastics are single-use products with a short lifespan (a few months for mulching films) (Espí et al., 2006; FAO, 2021), creating a disposal problem. Even though those plastics are made from recyclable materials, their use often contaminates them with soil and organic matter, complicating their recyclability (Agriculture Plastics Environment, 2021). On some occasions, alternatives to agricultural plastic disposal include burning, burying, or dumping in landfills (Orzolek, 2017). However, because of its resistance (FAO, 2021), susceptibility to weathering, and incomplete recovery after harvesting, residues of different sizes can enter terrestrial ecosystems and persist, harming the environment in different ways (FAO, 2021).

Once plastic residues accumulate in soil, they can alter soil physical and chemical properties. Many reviews have already summarised the effects that plastic residues, especially microplastics, will have on soil ecosystems (Chia et al., 2022; Kumar et al., 2020; R. Qi et al., 2020; Steinmetz et al., 2016). Some of the influences are changes in the soil porosity, bulk density, water movement (e.g., infiltration), soil pH, soil structure, interaction with soil components (e.g., soil aggregates), and release of chemicals (e.g., additives) (Boots et al., 2019; Chia et al., 2022; Pereira et al., 2021; Y. Qi et al., 2020). Moreover, plastic surfaces can act as vectors for the adsorption of chemicals and heavy metals (Chia et al., 2022; Moeck et al., 2022). Because of all these influences, plastic residues can disrupt soil fauna and microbial communities, enzymatic activities, seed germination, and nutrient cycling (Macan et al., 2024; McKay et al., 2022; Pereira et al., 2021; Zhao et al., 2022). Overall, these side effects can affect crop productivity.

1.4 From films to fragments: fragmentation and degradation processes

That plastic residues are accumulating everywhere is beyond doubt (see more in Chapter 3). But how exactly is this happening? Exposure to weathering conditions, such as heat, humidity, and solar radiation, can affect the stability of plastic materials, causing, for instance, embrittlement, discolouration, loss of elasticity and strength, and reduced impact resistance (Massey, 2007). Fragmentation into smaller sizes is often linked to different degradation processes and weathering conditions. Mechanical stresses, such as wind abrasion, tillage, and handling, accelerate fragmentation (Ren & Ni, 2022; Song et al., 2017), frequently removing degraded layers and exposing undegraded layers to weathering (see more in Chapter 2).

Photodegradation is one type of degradation that agricultural plastic can undergo in the environment. In brief, when the plastic absorbs a photon of light, most often ultraviolet radiation, the absorbed energy can cause chain scission (de Paoli, 2008; Verdu, 2012), thereby fragmenting the plastic. However, the degradation of agricultural plastics is far more complex, as different factors, such as additives, heat, humidity, and abrasion, can influence how and at what rate these materials degrade and fragment in the environment (Chamas et al., 2020).

A sustainable mitigation strategy for persistent plastic residues in agricultural soils is the development of soil-biodegradable mulch films. They are certified under EN 17033 (2018) to achieve at least 90% mineralisation to CO_2 in soil within two years in laboratory conditions. Biodegradable films are made from natural-based polymers, such as starch, and synthetic biodegradable polymers, such as polylactic acid or poly(butylene adipate-co-terephthalate) (Gao et al., 2021). Due to their biodegradability, these kinds of mulch films are tilled into the plough layer after use, eliminating the need for removal and disposal (Sander et al., 2023).

However, field studies show that soil-biodegradable residues can persist for several years (Ghimire et al., 2020; Sintim et al., 2020), suggesting accumulation of residues in soil with repeated use. Soil-biodegradable mulch films are certified to biodegrade in agricultural ecosystems; however, if they end up in a different environment, such as an aquatic environment, the certification is no longer valid, and they may persist in the receptor environment (SAPEA, 2020; van Grinsven & Schubert, 2023). If soil-biodegradable plastics persist in the environment, the risks they pose are likely to be the same as those of conventional non-biodegradable plastics (SAPEA, 2020).

1.5 Transport of plastic residues via rainfall influence

Agricultural soils are recognised as a potential major sink of plastic residues originating from multiple sources (R. Qi et al., 2020). Differences in regional climate, crop type, and management practices, such as the use of compost, sewage sludge, irrigation, and fertilisers, can also have a considerable influence on the concentration of plastic residues in agricultural soils (Bläsing & Amelung, 2018; Ding et al., 2020; Haixin et al., 2022; Ren et al., 2023; Rillig &

Lehmann, 2020; Sa'adu & Farsang, 2023; Stefano & Pleissner, 2022; Weber et al., 2022). Improper waste disposal, tyre wear, and atmospheric deposition are also contributing factors to the accumulation of plastic residues (Bläsing & Amelung, 2018; Maqbool et al., 2023; Piehl et al., 2018; Rehm & Fiener, 2024; Ren et al., 2023; Stefano & Pleissner, 2022).

Rainfall-induced surface runoff and erosion have been identified as a key pathway linking terrestrial plastic contamination to surrounding environments (see more in Chapter 4), such as the aquatic environment (Han et al., 2022; Koutnik et al., 2021; Lwanga et al., 2022; Rehm et al., 2021; Severe et al., 2025). Rainfall simulation studies have shown that plastic residues (e.g., microplastics) initially present at the soil surface can be exported during runoff events (Crossman et al., 2020; Han et al., 2022; Klaus et al., 2024; Severe et al., 2025; Zhang et al., 2022). On the one hand, changes in the physicochemical properties (e.g., size, shape, and functional groups, caused by environmental factors) (see more in Chapter 2), can increase hydrophilicity and modify surface charge, increasing their mobility (Liu et al., 2019). Furthermore, degraded plastic residues can reduce aggregate stability and affect erosion resistance (Dong et al., 2021; X. Li et al., 2024). On the other hand, however, microplastics may become attached to heavier soil particles or become embedded in soil aggregates, which reduces their tendency to be transported (Rehm et al., 2021).

1.6 Analytical and methodological challenges

Plastics vary considerably between types and forms (Shim et al., 2017; Wang et al., 2024). This variability normally limits the detection of these particles in different soil matrices. To overcome the diversity of plastic characteristics and complexity of the matrices of soil, the combination of different techniques is applied to increase the recovery of plastic residues and reliability of studies (e.g., different density separation, digestion, and microscope methods) (Lodh et al., 2025; Shim et al., 2017; Wang et al., 2024). However, all used methods have advantages and disadvantages (Lodh et al., 2025; Shim et al., 2017). Consequently, the use of different detection methods is often driven by the suitability for a certain research question, making comparability across studies difficult (Shim et al., 2017; Wang et al., 2024).

Wang et al. (2024) compared five detection methods for the same plastic particles, with different sizes, in the same soil matrix and concluded that the methods differed in accuracy across different size ranges of the same plastic material. Similarly, Peneva et al. (2025) compared several analytical methods for quantifying eight types of microplastics in three soils. Concluding that different methods exhibit distinct sensitivities to different microplastics across different soil matrices. Hurley et al. (2018) also tested different approaches to extract microplastics from organic-rich matrices and found that different protocols will affect the physical, even the chemical, integrity of certain polymers.

Besides variability in analytical methods, regional climates, sampling techniques, proximity to different input sources, incomplete information on quality assurance/quality control, and management practices can also increase variability in the final data (Bläsing &

Amelung, 2018; Lodh et al., 2025). Even differences in size limits and the final units in which the data are reported (Bläsing & Amelung, 2018) make data comparison across studies difficult (Shim et al., 2017). Koutnik et al. (2021) analysed microplastic concentrations reported in 196 studies from 49 countries, concluding that the concentration in soil and water bodies can vary up to eight orders of magnitude depending on the location. For instance, the authors attribute the variation in agricultural soils to the soil sampling method (e.g., sampling depth, number of samples, and soil processing), analytical methodology, and size range classification of the plastic residues (Koutnik et al., 2021).

Although macroplastics present the possibility of being detected and sorted visually (Berenstein et al., 2024; Li et al., 2022; Maqbool et al., 2024; Piehl et al., 2018), they are still understudied compared to microplastics (Maqbool et al., 2024). However, even though they can be visually identified, they are prone to misclassification, as natural fragments may look similar to synthetic ones (Piehl et al., 2018), requiring additional validation techniques.

Additionally, since macroplastics generally occur at lower numbers than microplastics, a better sampling strategy is needed for macroplastic studies. Moreover, the heterogeneous spatial distribution of macroplastic concentration (Weber et al., 2022) increases the need for a larger soil sampling to account for variabilities and ensure representativeness (Yu & Flury, 2021).

1.7 General objectives, general hypotheses, and structure of the thesis

Due to their agricultural usefulness, films made from various plastics are indispensable in modern agriculture. In Europe, especially, a year-round vegetable supply would be hardly possible without the extensive use of plastic films.

Creating a completely closed recycling loop isn't possible, and since even soil-biodegradable films only break down in the soil over time and under specific conditions, it's inevitable that small film residues will accumulate in the soil. We're still learning about how quickly these residues break apart, how much plastic is present at any given time, and how this influences whether the soil becomes a storage place or a source of film residues that might move to other ecosystems.

The overarching aim of this thesis is to shed light on the processes and interactions involved in plastic film fragmentation, accumulation in soil, and possible loss from soil via lateral transport pathways. The general objectives of the thesis are:

(i) To characterise and understand the fragmentation behaviour of conventional agricultural plastics under relevant environmental stressors (ultraviolet radiation, mechanical abrasion, and their interaction) in order to identify the mechanistic pathways through which macro- and microplastic residues are generated in agricultural settings.

(ii) To explore the accumulation of conventional mulch film-based plastic residues after decades of different plastic use, soil management, climate, and removal efforts in different regions of Europe.

(iii) To analyse the soil physical or soil hydrological effects of mulch film residues upon surface runoff and soil erosion, which potentially accelerates the loss of especially small plastic residues to neighbouring ecosystems. Compared to the first objectives, this is analysed in the case of soil-biodegradable mulch films intentionally ploughed into soil after use.

The thesis will verify or falsify the following general hypotheses, which will be specified in more detail in the central chapters of this work:

(i) The interaction between ultraviolet radiation and mechanical abrasion will accelerate degradation and formation of microplastics.

(ii) Different climate and management practices will influence plastic residue concentration.

(iii) Plastic residues incorporated in the soil will influence surface runoff and soil erosion, as well as the transport of residues to surrounding ecosystems.

The thesis focuses on the lifecycle of agricultural plastics, connecting their degradation, accumulation, and transport behaviour. Chapter 2 begins with a lab-scale experiment conducted under controlled conditions to enlighten our understanding of the degradation of agricultural films. After understanding how the degradation of agricultural plastics behaves, Chapter 3 investigates, on a field-scale experiment, other variables affecting plastic residue accumulation in agricultural soils. Finally, Chapter 4 evaluates the environmental consequences of agricultural plastic use via rainfall experiments to address the potential transport of plastic residues to surrounding ecosystems and their influence on soil processes. Figure 1.2 illustrates the outline of this thesis and highlights the interconnections among its chapters.

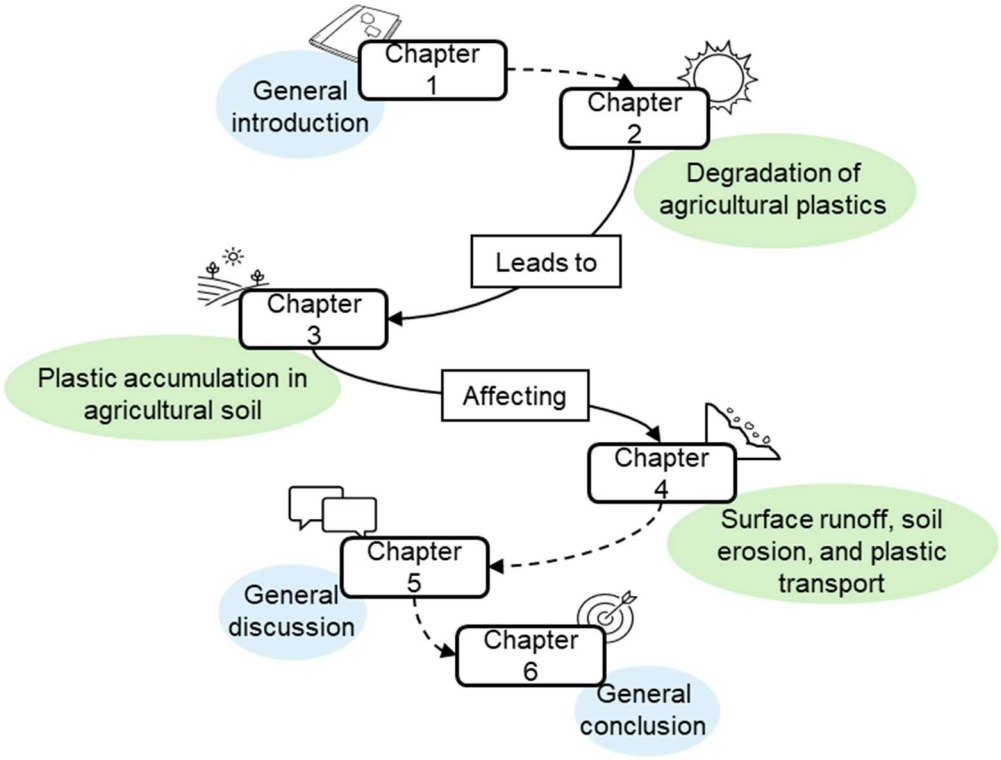

Figure 1.2: Overview of the thesis outline.

UV light and abrasion's role in degrading plasticulture films[1]

2

2.1 Introduction

The advent of plastic has revolutionised many sectors, including agriculture. The use of plastic in agriculture, known as plasticulture, started in the early 1950s with low-density polyethylene (LDPE) as mulch film and, since then, has been steadily increasing (Espí et al., 2006). A large variety of plastics are used in agriculture, depending on the purposes, including different types of plastic films (Agriculture Plastics Environment, 2019; Bläsing & Amelung, 2018), irrigation systems, nets, transport, and packaging of agricultural food products and chemicals (FAO, 2021). In modern horticulture, plastic films are used to cover the soil (i.e., mulch films) or the crop (i.e., tunnel and greenhouse films). Because of their crucial role, plastic films are the most commonly used plastic in vegetable production, with mulching films and greenhouse accounting for 83×10^6 and 120×10^6 kg applied in Europe's agriculture, respectively (Agriculture Plastics Environment, 2019). Depending on the type of film, plasticulture can improve growing conditions by limiting evaporation, regulating temperature, increasing or decreasing sunlight absorption, suppressing weeds, or benefiting soil sterilisation (Hann et al., 2021; Steinmetz et al., 2016). To tailor them for their intended use, plastics are modified by chemical additives such as fillers, antioxidants, and stabilisers (Steinmetz et al., 2016; Tang et al., 2023), affecting specific properties such as elasticity, strength, thermal and ultraviolet (UV)-radiation stability, and colour (Hann et al., 2021; Steinmetz et al., 2016). However, plastic film producers and vendors hardly provide any details regarding the use of additives. Plastic films are also produced in different thicknesses (8 to 300 µm), and can be separated into thin (< 80 µm) and thick (> 80 µm) films. Thin films are used as mulching for one growing season, while thick films are primarily used in horticulture, vegetable, or fruit production, where plastic films are used for multiple cropping seasons and are supposed to remain stable for a longer application time.

Despite the advantages introduced in agricultural production, weathering and mechanical stress may lead to the detachment of residues during mulch application or at the time of removal, especially when films approach the end of their intended lifespan (Ren et al., 2021; Zhang et al., 2021). This process can either lead to the formation of macroplastic (> 5 mm), which is subsequently fragmented into smaller pieces, particularly during tillage operations (Maqbool et al., 2024), or to the direct formation of secondary microplastic particles (< 5 mm) via abrasion (Ren et al., 2020). Hann et al. (2021) estimated that if at least 5% of the 83×10^6 kg of mulch film annually applied in Europe are not removed after use, 4.15×10^6 kg would remain in the soil system. Plastic residues have been shown to impact soil systems in

[1] An adapted version of this chapter was published at PlosOne as: Cugler Moreira AC, Wilken F, Fabrizi A, Fiener P (2026) UV light and abrasion's role in degrading plasticulture films. PLoS One 21(3): e0344790. https://doi.org/10.1371/journal.pone.0344790.

terms of physical and chemical properties, with a pronounced effect on the soil biome, reducing plant growth and, hence, agricultural productivity (Boots et al., 2019; R. Qi et al., 2020). Transport of plastic residues via wind (Rezaei et al., 2022), surface runoff, and soil erosion (Rehm et al., 2021) connects agricultural plastic inputs with neighbouring ecosystems. Moreover, small microplastic particles may be transported vertically along the soil column (W. Li et al., 2024) and hence result in the contamination of shallow groundwater (W. Li et al., 2024; Ren et al., 2021). Yang et al. (2022) studied the microplastic formation from biodegradable, oxo-degradable, and non-degradable agricultural plastic films under different UV irradiation exposures in soil. Non-degradable films generated fewer microplastic particles, and crystallinity and abrasion were identified as key accelerating processes. Song et al. (2017) found that LDPE pellets fragmented more under combined UV and abrasion exposure than with abrasion alone. Bhattacharjee et al. (2023) also reported that non-agricultural LDPE films release more microplastics through abrasive forces and photodegradation exposure compared to pristine films. In contrast, Ren and Ni (2022) questioned the primacy of photodegradation in microplastic formation of transparent agricultural LDPE film, suggesting wind-driven abrasion as a more significant contributor. To mitigate the plastic input to soil systems, improvements in the process of understanding the weathering and fragmentation pathways of different plastic films in agricultural environments are therefore essential (Ouyang et al., 2023; Ren & Ni, 2022; Yang et al., 2022).

This study aims to get a mechanistic understanding of photodegradation and follow-up abrasion using state-of-the-art analytical methods. The research questions of this study are: (i) What is the temporal development and degree of photodegradation at different UV radiation exposure times for different types of films used in plasticulture? (ii) How does this photodegradation correspond to the formation of microplastics and macroplastics? (iii) What is the role of plastic film abrasion in degradation and for plastic input into soil systems?

2.2 Materials and methods

2.2.1 Experimental design

In this experiment, commercially available LDPE films were used (Firmenich, Germany), which are commonly applied as short-term (one season), long-term mulch films (multiple seasons), and greenhouse covers (multiple seasons). Two thin black mulch films (approx. 20 µm), two thick black and white mulch films (approx. 100–150 µm), and two thick transparent greenhouse covers (approx. 150–180 µm) were purchased at two different big farm suppliers in 2022 (Table 2.1).

Table 2.1: Main characteristics and typical low-density polyethylene (LDPE) film usage.

Abbreviation‡	Measured thickness* (µm)	Type	Use	Duration use	Life span†	Colour
MS1	17.6 ± 1.8	thin mulch film on the soil surface	vegetables or fruits	one growing season	up to 36 months	black
MS2	18.4 ± 1.0					
ML1B	98.9 ± 2.7	thick mulch film on the soil surface	asparagus	multiple growing seasons	up to 84 months	black/white
ML1W						
ML2B	143.4 ± 4.9					
ML2W						
GL1	156.8 ± 1.4	large tunnel film for greenhouses	vegetables or fruits	Multiple growing seasons	up to 54 months	transparent
GL2	185.3 ± 5.9					

‡ MS1: mulch film, short term, no. 1; ML1B: mulch film, long term, no. 1, black side up; ML1W: mulch film, long term, no. 1, white side up; GL1: greenhouse film, long term, no. 1; * mean thickness ± standard deviation measured at ten random points on a sample with an approximately 20 cm x 20 cm size; † manufacturer information.

For the experiment, an approximately 1 m long part of the mulch and greenhouse films were withdrawn from each specific roll. From each of the 1 m long samples, twenty 3 cm x 5 cm small samples (160 samples in total) were taken out. The samples were separated into 72 samples for different UV treatments, 72 samples for different UV plus abrasion treatments, 8 samples without UV but with abrasion treatment, and 8 samples representing pristine films as controls (Figure 2.1). Each 3 cm x 5 cm piece was placed on a magnetic frame with an internal window size of 2 cm x 4 cm and an external frame size of 4 cm x 5 cm to keep the samples stable and as flat as possible during the UV treatment.

For the UV treatment (Figure 2.1), a UV xenon test chamber (Q-SUN Xe-1-SE, Q-Lab, USA) equipped with one xenon arc lamp 1800 W (X-1800+, Q-Lab, USA), a Daylight-Q filter, and an insulated black panel temperature sensor was used. In terms of comparability between the artificial UV radiation source and natural sunlight exposure, the test chamber matches the ASTM G155 standard (ASTM, 2021). The UV test chamber was used with a spectral range from 300 to 400 nm (mainly UVA), irradiation of 75 W · m^{-2} (where this represents 6.48 MJ · m^{-2} for one day under UV exposure), and a chamber temperature of 50°C in a 24-hour light cycle. No other parameter was controlled. The time the samples spent in the chamber was set to simulate nine semi-randomised UV exposure times (21, 26, 31, 39, 40, 42, 52, 60, and 80 UV days), reflecting the variety of UV exposure conditions found in Europe. The samples were stored at 4°C between treatments and analysis to prevent further degradation.

Following the European organisation for technical approvals (2004), an estimated recommendation of 201 MJ · m^{-2} was established for one year of equivalent radiation dose for the wavelength range between 300–400 nm. To determine the relative exposure of the samples to the natural conditions, the incident energy per day (6.48 MJ · m^{-2}) should be multiplied by the number of exposure days and then divided by the one-year recommendation (201 MJ · m^{-2}).

Due to the limited number of samples, after the UV treatment, one sample of each mulch film was divided into two 2 cm x 2 cm pieces. One half was first used to perform the roughness test, then treated with mechanical abrasion and finally analysed for roughness, wettability, and chemical changes. The second half was only UV-treated and used to check the mechanical properties. The second sample of each mulch film kept its original size (2 cm x 4 cm); however, one-half was used to check for chemical change, and the other half was used for wettability change. The mechanical abrasion treatment was designed to simulate the potential mechanical abrasion that may occur due to wind-induced movement of the film on soil (particularly important for mulch films) and the possible impact of soil particles moved with the wind over the film surface (Figure 2.1). Therefore, the samples were mixed with 20 g of standard loamy sand soil (LUFA Speyer, 3.6 ± 1% of clay, 8.6 ± 1% of silty, 87.8 ± 1% of sand) in a 60 mL amber screw neck glass vessel. The vessels were placed in a roller mixer (RG11, Roller Grill, France) with 4 rpm velocity for 61 days, with no replicates. The abrasion treatment was carried out in dry conditions at room temperature, without temperature control. After the abrasion exposure and before the material analysis, all abrasion samples were washed in an ultrasonic bath (130/300 W, 40 kHz) for 5 minutes using deionised water to remove excess soil particles.

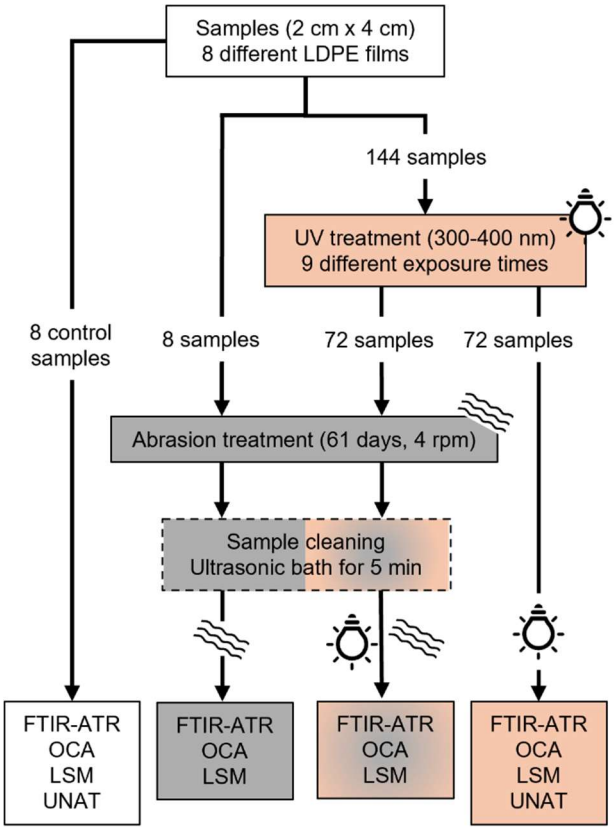

Figure 2.1: Scheme of the sample treatment and analyses.

2.2.2 Analysing film properties

2.2.2.1 Fourier transform infrared with attenuated total reflectance (FTIR-ATR)

FTIR-ATR was used to evaluate the chemical changes after the treatments for all samples. The technique is sensitive to changes in the polymer chemical structure and the detection of polar groups, such as C=O (Chamas et al., 2020). The chemical change was determined using an FTIR-ATR spectroscopy (Equinox 55, Bruker, USA) equipped with an ATR diamond crystal accessory with a penetration depth of around 2 µm.

Degradation was quantified by calculating the carbonyl index (CI) and the hydroxyl index (HI), which quantify the increase in carbonyl and hydroxyl groups, respectively, compared to a reference band. CI and HI were determined by dividing the area of the carbonyl band ($A_{carbonyl\ band}$), or the hydroxyl band, by the area of the reference band ($A_{reference\ band}$) (equation 2.1) (Almond et al., 2020; Chamas et al., 2020; Hirsch et al., 2018; Rouillon et al., 2016; Zhang et al., 2021). In addition to the area-based method, CI and HI were also calculated using the peak height values of the carbonyl (Figure S1) and hydroxyl (Figure S2) bands relative to a reference peak, allowing for a complementary evaluation of degradation. To calculate CI and HI, the wavelength range, as the wavenumber peaks, for the carbonyl, hydroxyl, and reference bands, was visually determined as given in Table 2.2.

$$CI = \frac{A_{carbonyl\ band}}{A_{reference\ band}} \tag{2.1}$$

Each spectrum was collected in transmittance mode, from 4000 to 400 cm^{-1} spectral range, with 32 co-added scans per spectrum and 4 cm^{-1} resolution. For each sample, three spectra were measured from random locations, while two spectra were collected on ML2B (treated with 52 days of UV exposure and abrasion) and MS1, MS2, ML1B, ML2B, ML1W, ML2W, and GL1 (treated with 80 days of UV exposure).

Table 2.2: Carbonyl, hydroxyl, reference band range, and wavenumber peak used to calculate the carbonyl and hydroxyl indices.

Sample	Bands (cm^{-1})			Peak (cm^{-1})		
	Carbonyl	Hydroxyl	Reference	Carbonyl	Hydroxyl	Reference
ML1B	1840–1480	3730–2980	740–650	1536	3330	717
ML2B	1840–1480	3730–2980	740–650	1536	3330	717
ML1W	1775–1510	3730–3110	1510–1390	1733	3400	1462
ML2W	1775–1510	3730–3110	1510–1390	1733	3400	1462
MS1	1840–1480	3660–3000	750–640	1712	3370	719
MS2	1840–1480	3660–3000	750–640	1712	3370	719
GL1	1840–1510	3730–3120	760–650	1570	3400	719
GL2	1840–1510	3730–3120	760–650	1570	3400	719

2.2.2.2 Optical contact angle (OCA)

An OCA instrument (OCA35, DataPhysics, Germany) was used to measure the samples' wettability changes in sessile mode. Wettability usually measures the interaction between a solid and a liquid, where small angles (< 90°C) refer to high wettability (hydrophilic) and high angles (> 90°C) refer to low wettability (hydrophobic) (Yuan & Lee, 2013). The contact angle technique can detect the formation of polar groups on the surface of the photodegraded sample, which leads to an uneven charge distribution of the plastic surface that causes measurable differences in the interactions with water (Chamas et al., 2020). A 2 µL droplet of distilled water was applied at room temperature. Ten seconds after the application, the contact angle between the film sample and the water drop was measured 9 times. Each sample was measured at three randomly selected locations over the sample (3 locations 9 times, totalling 27 measurements).

2.2.2.3 3D laser scanning confocal microscope (LSM)

The surface roughness of all samples was measured using an LSM (VK-X1000 Series, Keyence, Japan). The LSM was equipped with a 404 nm laser, and a confocal light was used to scan the sample surface before and after each test. The samples were placed on the same magnetic frame used for UV radiation exposure. Subsequently, high-resolution scans of the films were carried out with 500 µm magnification, obtaining for each sample a 1024 x 768 raster with 2.7 µm x 2.7 µm cell size. The terrain ruggedness index (TRI) was calculated using the rasters in R software ('tri' function, 'spatialEco' library) to evaluate the mean small-scale roughness changes. The TRI function calculates the sum of the absolute differences in elevation between a specific raster cell and its surrounding cells (Riley et al., 1999). In order to avoid smoothing effects by averaging over a large area, only the direct neighbouring cells of a 3 x 3 raster window were taken into account for the calculation of the TRI. Finally, the average TRI and its standard deviation were calculated over the entire area of each sample. Thereby, average surface roughness was determined using LSM, which integrates roughness across the full sample area to yield a representative mean for each treatment. This representative mean was prioritised over additional replicates of smaller areas.

2.2.2.4 Universal nanomechanical tester (UNAT)

To measure changes in mechanical properties between pristine and UV-degraded samples, the variation in Young's modulus was recorded using a UNAT (ASMEC, Zwick/Roell, Germany). The UNAT applies controlled forces using a Berkovich indenter and records the material response, which allows for LDPE statements about crystallinity, brittleness, and elasticity (Rabek, 1995). The measurement is structured by gradually applying a force using a Berkovic indenter of 10 mN over 10 seconds (loading phase), maintaining this force for 5 seconds (holding phase), and then reducing the force to 0.260 mN over 4 seconds (unloading phase), with a fixed Poisson's ratio of 0.4. This procedure records the penetration depth and elastic and plastic deformation, allowing the obtention of sample elasticity (Bairral, 2012). We chose a subset of UV exposure encompassing the entire degradation spectrum of the different

film types. The UV plus abrasion-treated samples were excluded from the analysis due to potential interference of soil particle attachment on the film surface. The following samples – MS2, ML1B, ML1W, and GL1, each subjected to UV treatment for 0, 21, 40, and 80 days – were selected and analysed by UNAT. The 0.5 cm x 1.5 cm samples were attached to glass slides with double-sided tape. The measurements were carried out 27 times at 3 different locations (position was randomly selected) on the sample. Within each location, 9 replicate measurements were conducted at close distances.

2.2.2.5 Statistical analysis

All the data representing the different treatments, measurements and their residual values were tested for normality using a Kolmogorov-Smirnov normality test (Kotz & Johnson, 1992). The samples analysed via FTIR-ATR and UNAT presented normally distributed data, the samples analysed via OCA presented a mixed distribution, and the samples analysed via LSM couldn't perform a normality test due to the lack of replicates. A regression line was performed to check for trends, an ANOVA test for the parametric data, and a Kruskal-Wallis test for the non-parametric data were performed to check for significant differences between treatments, using a significance level of $p < 0.05$ for all statistical tests. All statistical analyses and graphs were done using OriginPro 2023b (OriginLab, USA).

2.3 Results

2.3.1 Spectral characteristic and chemical groups

The spectral difference between the sample treated for 80 days and the pristine sample (Δ spectra) identifies the changes caused by the treatments (Figure 2.2). Although the Δ spectra highlight notable changes in the regions of 3100–3700 cm^{-1} and 910–1080 cm^{-1} connected to the abrasion treatment (Figure 2.2b) (Ge et al., 2014; Smidt & Meissl, 2007), the figure may not be suitable for illustrating trends over time. As such, a dedicated index (i.e., carbonyl index) remains necessary to assess degradation over time and treatment.

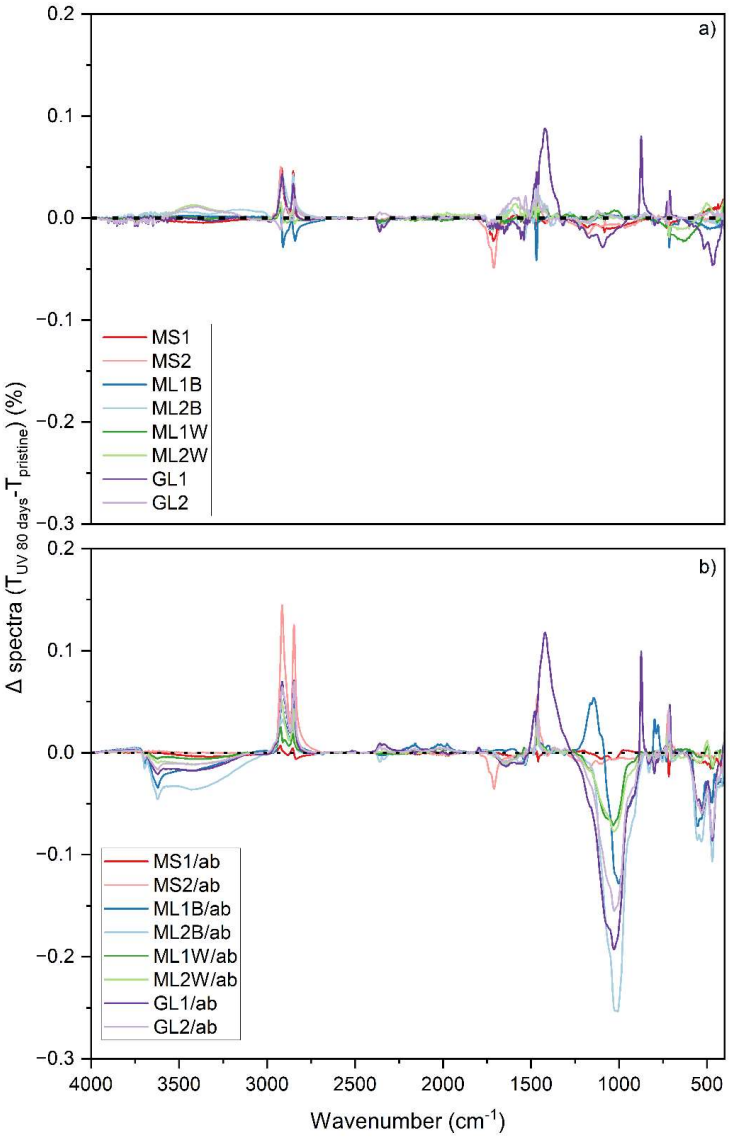

Figure 2.2: Δ spectra between 80 days UV-exposed and pristine samples. The different colours indicate the mean Δ spectra of the different samples tested in this study. Where a) represents the samples treated only with UV, and b) represents the samples treated with UV and abrasion. For an overview of the abbreviation, please see Table 2.1.

The chemical analysis using the carbonyl index (area under the curve method) revealed no significant changes in the surface properties of the films subjected to both UV treatment and abrasion (Figure 2.3). This indicates that, for the majority of the samples, these treatments did not alter the chemical composition of the film surfaces. However, an exception was observed with the film sample MS2. This particular sample exhibited a significant change in its surface properties, demonstrating a clear linear relationship with the carbonyl index values (r^2 = 0.80 and 0.74). This suggests that the MS2 film is more susceptible to degradation under the combined effects of UV treatment and abrasion compared to the other samples. While for the height values, a significant change was observed for the samples MS2 (r^2 = 0.69 for the UV-treated sample, r^2 = 0.74 for the UV plus abrasion-treated sample), ML1B (r^2 = 0.51 for the UV-treated sample), ML2B (r^2 = 0.51 for the

UV-treated sample), GL1 (r^2 = 0.49 for the UV-treated sample), and GL2 (r^2 = 0.40 for the UV plus abrasion-treated sample) (Figure S1).

The ANOVA test, for both methods, showed significant differences ($p < 0.05$) between the two treatments (UV vs. UV plus abrasion) for all samples. All samples showed significant differences when comparing different samples (e.g., MS1 and GL2) under the same treatment.

Due to interferences caused by the abrasion test in the hydroxyl region (see Figure 2.2 and Table 2.2), the hydroxyl index using both methods (area under the curve (Figure S3) and height value (Figure S2)) was performed only for the UV-treated samples. However, none of the samples presented any significant change or linear trend.

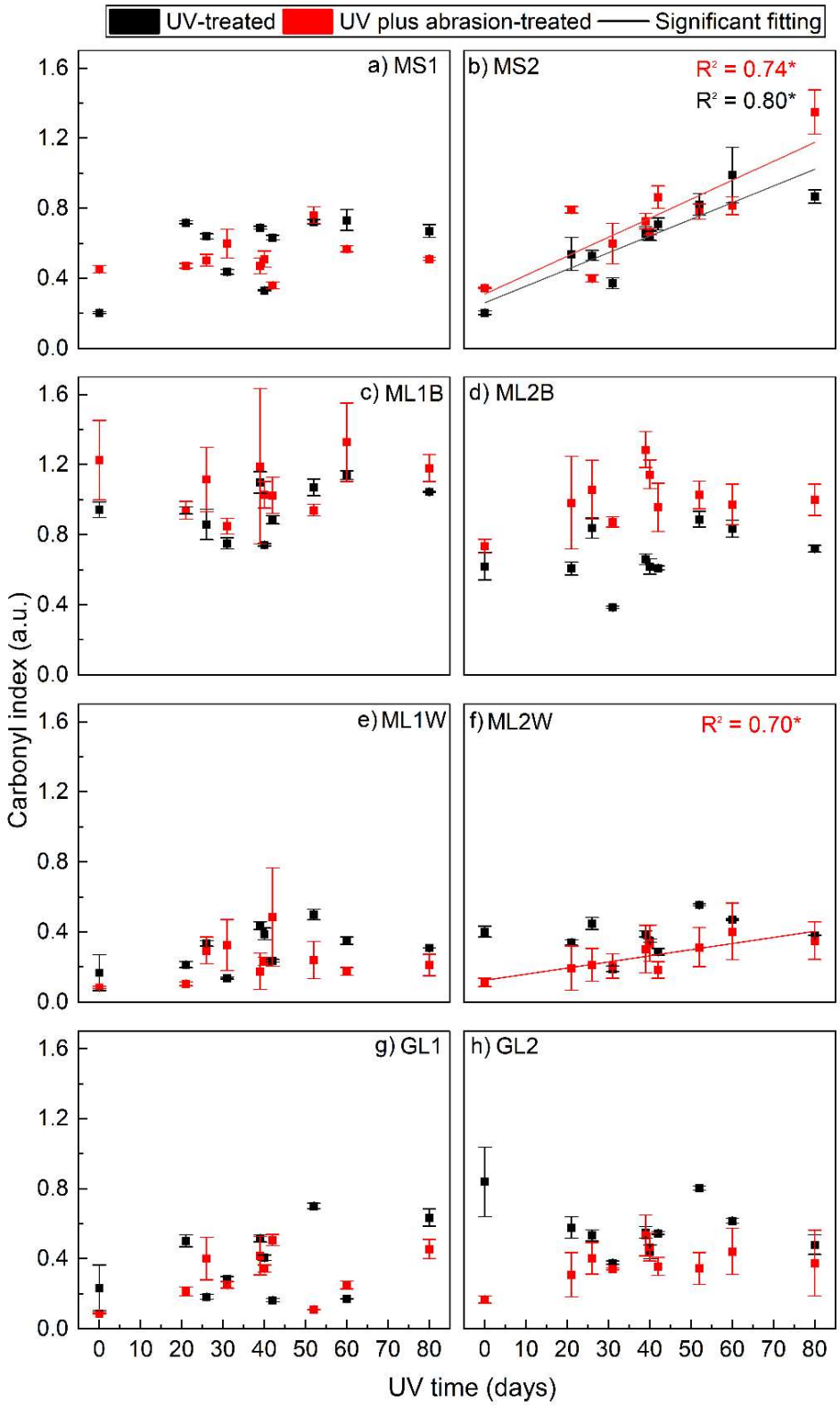

Figure 2.3: Carbonyl index of a) MS1, b) MS2, c) ML1B, d) ML2B, e) ML1W, f) ML2W, g) GL1, and h) GL2. *p < 0.05. For abbreviations, see Table 2.1.

2.3.2 Wettability

The contact angle analysis of water droplets on the film surface (wettability) demonstrated a significant trend with UV exposure time for all UV-treated samples (Figure 2.4).

The wettability of these samples increased rapidly from the pristine state to the UV-exposed state within the first 21 days of UV treatment. Following this initial surge, the rate of change in wettability slowed down for the remainder of the UV exposure period. The relationship between UV exposure time and wettability was exponential, with an r^2 value of at least 0.58. In contrast, the majority of samples treated with both UV and abrasion exhibited much higher wettability, similar to that of the pristine film, with no significant changes observed over the UV exposure time (Figure 2.4). The Kruskal-Wallis test indicated significant differences between the same samples when treated with different methods (UV vs. UV plus abrasion). Furthermore, samples with the same intended use but different manufacturers also showed significant differences.

2.3.3 Surface roughness

The roughness analysis following UV treatment and abrasion revealed that almost all the UV plus abrasion-treated samples exhibited higher roughness compared to the samples treated only with UV, except for both greenhouse films (GL) samples (Figure 2.5g and h). Hardly any significant linear trend with UV radiation exposure time was observed for any of the samples except for the GL2 UV-treated sample (r^2 = 0.44; Figure 2.5). An ANOVA test was performed to check the significant differences between different treatments for the same samples, and only the sample ML2W did not present a significant difference. Comparing different samples submitted to the same treatment, all samples presented significant differences.

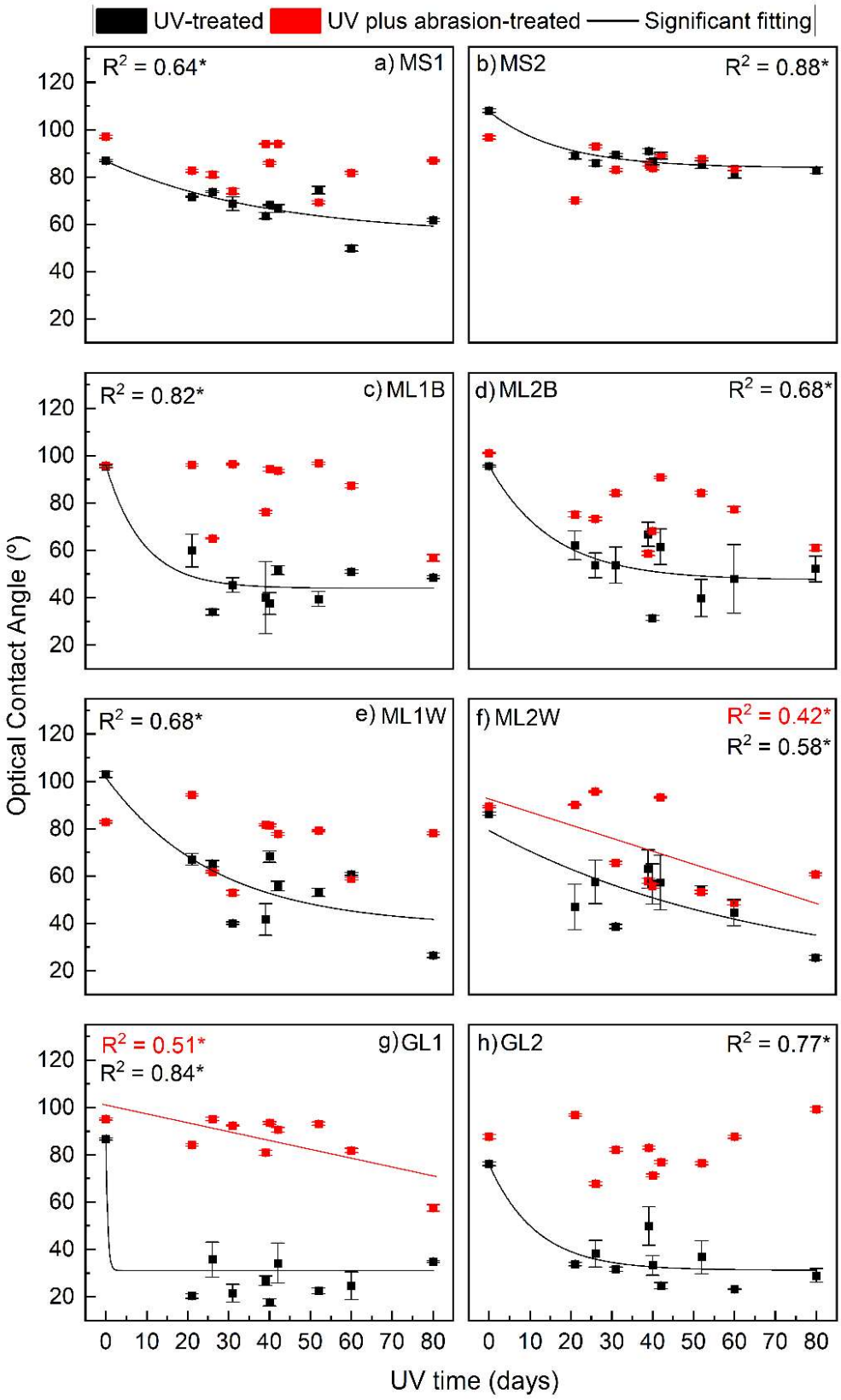

Figure 2.4: Wettability of a) MS1, b) MS2, c) ML1B, d) ML2B, e) ML1W, f) ML2W, g) GL1, and h) GL2. *$p < 0.05$. For abbreviations, see Table 2.1

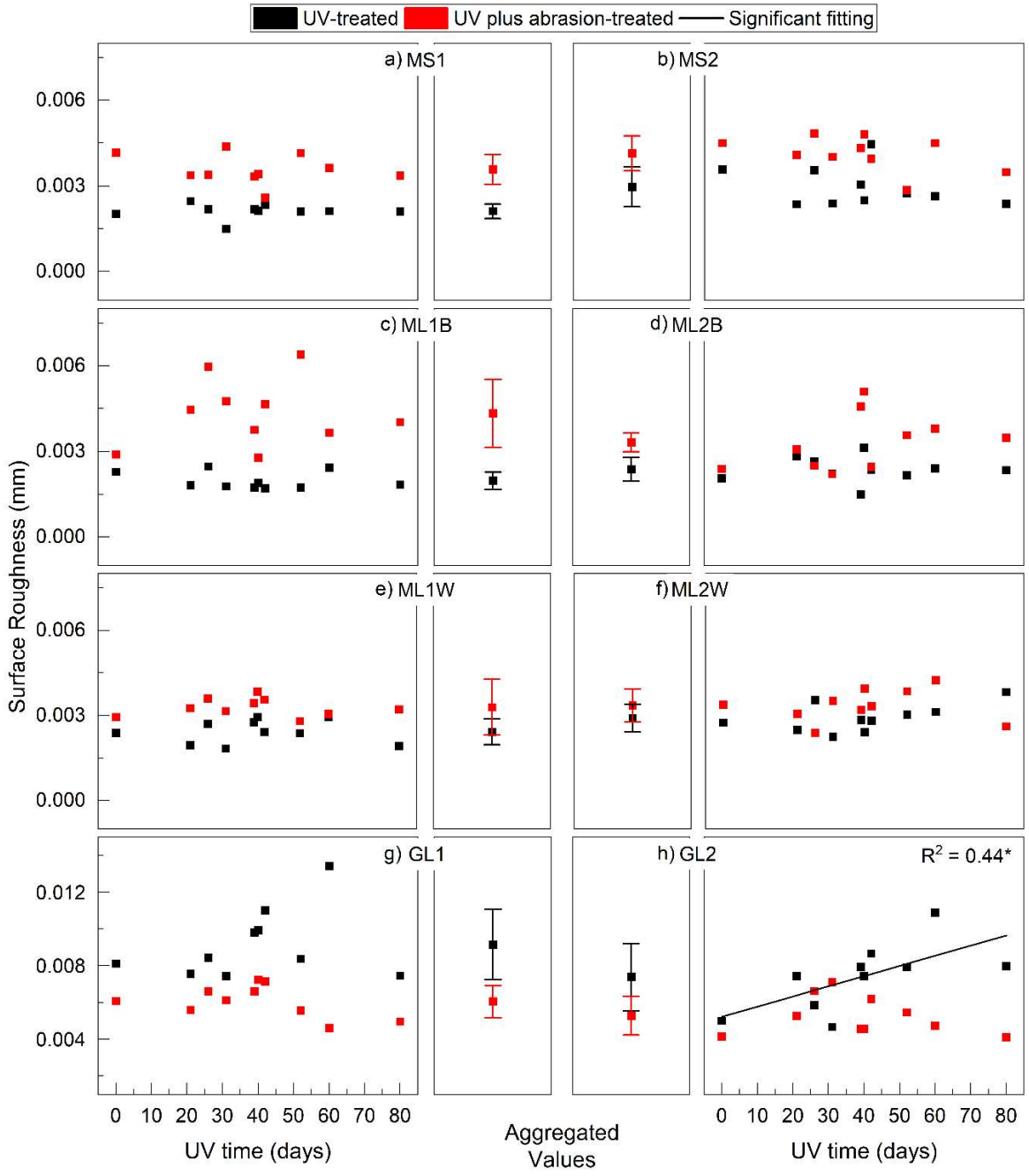

Figure 2.5: Surface roughness of a) MS1, b) MS2, c) ML1B, d) ML2B, e) ML1W, f) ML2W, g) GL1, and h) GL2. *p < 0.05. The samples GL1 and GL2 are presented in different scales for better visualisation. For abbreviations, see Table 2.1.

2.3.4 Elasticity

The elasticity test using the nanoindenter indicated no significant change in the Young's modulus (modulus of elasticity) for different UV radiation exposure amounts (Figure 2.6). However, was observed for the ML1B sample, a decrease from the pristine level to 21 days of UV exposure. It is important to note that the greenhouse film (GL1) exhibited much higher elasticity compared to the mulch films tested (ML1B and ML1W; Figure 2.6). The ANOVA test revealed that only the pristine ML1B sample showed a significant difference from the other ML1B samples tested. The changes in the elastic properties of the thin film sample (MS2) could not be measured due to technical limitations.

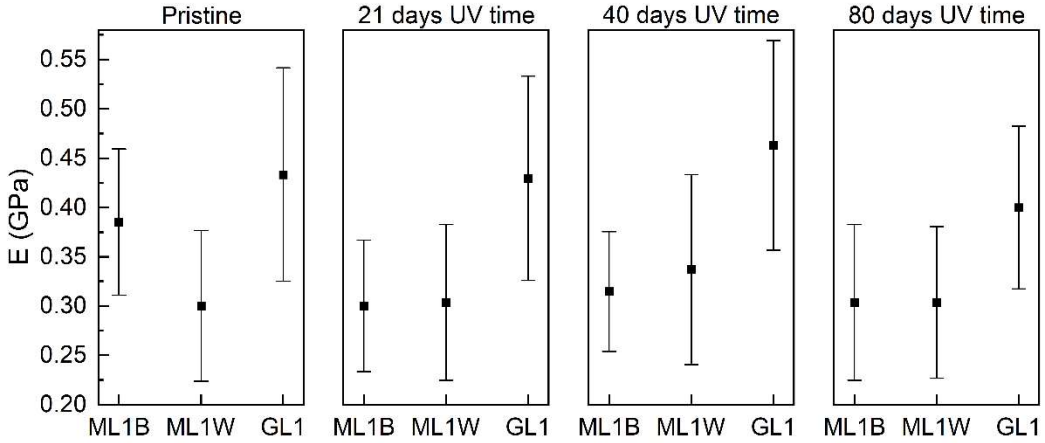

Figure 2.6: Young's modulus (E) of UV-treated samples. For abbreviations of the tested films, see Table 2.1.

2.4 Discussion

Various films used in plasticulture were exposed to different durations of UV radiation and to mechanical stress through mineral soil abrasion in this study. Subsequently, the films were assessed for changes in carbonyl and hydroxyl content, wettability, roughness, and elasticity. Even though two different methods for the degradation index are presented (area under the curve and height value), only the area under the curve method will be further discussed, as we believe this method increases the level of precision by considering all carbonyl species, series of overlaps, and band shifts present, avoiding error when choosing one specific peak for the height (Almond et al., 2020).

Both the FTIR-ATR-based index (carbonyl and hydroxyl) and the wettability of plastic samples are related to changes in polar groups (Chamas et al., 2020; Hirsch et al., 2018; Rouillon et al., 2016). Although the hydroxyl band could serve as an alternative assessment for degradation, the abrasion treatment introduced spectral interference in this region (Figure 2.2) (Ge et al., 2014). Since the abrasion test interference makes it difficult to accurately compare the UV-treated and UV and abrasion-treated samples, the hydroxyl index will not be further discussed.

Abrasion could have introduced cracks that allowed oxygen to diffuse into the sample, potentially leading to observable degradation (Figure 2.3). Alternatively, the abrasion process might have removed the outer surface layer, exposing a more preserved inner part of the samples (Figure 2.3). While the range of UV exposure carried out within this study did not yield a pronounced significant change in the carbonyl index for the majority of samples (Figure 2.3), the wettability (reduction of water droplet contact angle) of the samples showed a substantial increase after a short exposure time (Figure 2.4).

The wettability increased rapidly after the first period of UV radiation exposure, but showed very limited reaction to additional UV radiation. Hence, wettability changes after a short sunlight exposure time, indicating the rapid formation of polar groups, which still fall under the detection limit of the FTIR-ATR-based carbonyl index (Figure 2.3). Based on Rouillon et al. (2016), who used other analyses, mainly mechanical properties analyses, stated that the detection of early-stage degradation of polypropylene could be possible even before the detection of carbonyl by FTIR. When similar samples are exposed to UV influence, transparent samples are expected to present a higher effect by the UV treatment (transparent > white> black). However, it needs to be highlighted that sample MS2 (mulch film designed for short-term application) showed an exceptionally high photodegradation response according to the carbonyl index (80% and 74% explained variance) in comparison to the sample MS1. A likely explanation is a substantially different plastic composition between polymers and additives (e.g. UV stabilisers) (Tang et al., 2023), which vary greatly among different producers and products and is a closely guarded trade secret. The use of additives is crucial for the degradation processes. Nevertheless, the different concentrations and types of additives will have distinct effects, as the combination of different polymer types and additives may present different sensitivities, influencing the degradation mechanisms (Niu et al., 2025). As shown in this study, films with similar purposes may react differently (Figure 2.3). For practitioners, the formation of microplastics and, thereby, plastic input to their soil systems can hardly be estimated, even though the film stability is tested with respect to the UV exposure and application time (Ren et al., 2020).

Using wettability for quantifying polar groups can serve as an early detection of degradation (Hirsch et al., 2018). A large change in wettability by photodegradation was also observed by Suresh et al. (2011). Additionally, in line with our results, Liang et al. (2024) also reported higher wettability on samples aged under soil burial. While wettability is also sensitive to changes in surface roughness (Hirsch et al., 2018; Yuan & Lee, 2013), this study showed that surface roughness changed only when abrasion was applied (Figure 2.5). This indicates that UV radiation does not cause larger surface morphological changes, such as detachment of microplastics, that are detectable by the laser scanning microscope (2.7 µm x 2.7 µm raster size), while the abrasive force has sufficient energy to change the film surface morphology and thereby release plastic particles (Bhattacharjee et al., 2023; Ouyang et al., 2023; Song et al., 2017).

For all mulching films, the non-UV but abrasion-treated sample roughness is higher than the pristine film conditions (Figure 2.5), indicating that abrasion is not limited to degraded films. Interestingly, the pristine transparent greenhouse film (GL1 and GL2) is distinctively rougher compared to the mulch film and shows smoothening as a reaction to abrasion (Figure 2.5). Abrasion has been shown to increase the wettability of UV-treated films, which points to the removal of surface particles by abrasion. Hence, a destabilisation of the polymer structure by photodegradation makes films susceptible to detachment by abrasion (Bhattacharjee et al., 2023; Hirsch et al., 2018; Ouyang et al., 2023; Song et al., 2017; Yang et al., 2022). Given the limited penetration depth of UV radiation and the associated formation of polar groups, abrasion can act as an agent to expose undegraded polymers to the surface and UV radiation. Hence, UV light and abrasion are interacting processes that may accelerate microplastic formation (Bhattacharjee et al., 2023; Ouyang et al., 2023; Song et al., 2017), with thinner films presenting a higher degradation rate (Uzamurera et al., 2023; Xiong et al., 2023). Thereby, plasticultural regions where abrasion is a more frequent process, such as storm-prone regions, where phases of UV radiation exposure frequently alternate with abrasion, may experience accelerated microplastic formation as degraded polymers are removed from the surface and stop sheltering undegraded deeper-located polymers from UV radiation (Chamas et al., 2020; Zhang et al., 2021). However, no microplastic analysis was performed to quantify the formation of microplastics.

Our results show that photodegradation requires very few days of accelerated UV radiation exposure and could potentially form detachable microplastic particles that abrasion processes in the soil system can release. Reduced film elasticity increases fragmentation to soils either during the application of mulch and greenhouse film or during the removal process after the cropping season. The UV radiation exposure time applied within this study (max. 80 days) did not cause changes in the elasticity of the tested samples (Figure 2.6). Based on an estimated conversion from artificial UV exposure to natural UV exposure (see experimental design section), and only considering UV radiation influence, as weathering could be influenced by other factors such as temperature and humidity, all mulch films used in this study fall within the manufacturer's specified range of usability (Table 2.1). Indications for a loss of film stability were not identified after the applied UV radiation exposure time. However, the lack of substantial change in film elasticity in this study does not necessarily exclude fragmentation of mulch and greenhouse films over longer exposure times and/or combined environmental factors. It only highlights that the parameters used in this study were insufficient to cause fragmentation.

2.5 Conclusion

In this study, different mulch and greenhouse films commonly used in plasticulture were exposed to various amounts of UV radiation and mechanical abrasion in a laboratory experiment. Even though the UV radiation exposure time for all films was less than the designated lifetime indicated by the manufacturer, our results show clear evidence of photodegradation on the different agricultural plastics used in this study, as well as an

accelerating effect of abrasion on the degradation process. Additionally, we found indications of potential microplastic formation associated with abrasion processes. Mechanical abrasion of surface particles proved to be an efficient process for exposing non-degraded surfaces. These results suggest that the removal of surface particles by abrasion exposes undegraded film layers to UV radiation, effectively acting as a degradation accelerator. This study indicates that microplastic formation occurs almost immediately after application and that abrasion is an important modulator of the degradation rate. However, the films tested in our study showed no loss of elasticity and, therefore, no increase in fragmentation potential during agricultural management.

Future laboratory-based research should consider other environmental variables also important for degradation processes that were not considered in this study, such as humidity and temperature. A logical next step is to monitor UV-induced film degradation under real-world field conditions.

Macro- and microplastic concentration in agricultural soils accounting for different management and regional conditions

3

3.1 Introduction

Since the early 1950s, when low-density polyethene was first used as a mulch film in agriculture, the use of plastic in this sector has steadily increased (Espí et al., 2006). An amount of about 2.4×10^9 kg of plastic was used in European agriculture in 2022 (Plastics Europe, 2024a). Plastic films used for mulch films, greenhouses, and small tunnels (Agriculture Plastics Environment, 2019) are still a very important part of the agricultural plastic production, covering approximately 27×10^6 ha worldwide (Petrovich, 2019), and mulch films covering 4.6×10^5 ha in Europe (Steinmetz & Schroder, 2022). The use of mulch films is essential to many activities, as they can function as crop or soil covers, improve water use efficiency, regulate temperature, suppress weeds, and improve growing conditions (Gao et al., 2021; Hann et al., 2021; Steinmetz et al., 2022). However, the intensive use of these agricultural plastics is associated with several environmental issues.

When exposed to prolonged weathering and mechanical stress, agricultural plastics may fragment into macroplastics (> 5 mm), microplastics (1–5 mm), and/or nanoplastics (< 1 mm) (Chapter 2) (Berenstein et al., 2024; Maqbool et al., 2024; Ren et al., 2021; Zhang et al., 2021) and become a persistent pollutant in the environment. When accumulated in soil (Li et al., 2022), these plastic residues can compromise soil health by altering soil porosity (Jiang et al., 2017), deteriorating soil structure (R. Qi et al., 2020; Y. Qi et al., 2020), hindering water movement (Wen et al., 2022), stressing soil microbial communities (Boots et al., 2019; McKay et al., 2022), facilitating the leaching of compounds (Macan et al., 2024; McKay et al., 2022), and changing soil temperatures (Wen et al., 2023). Consequently, plant health and agricultural productivity are also affected (Boots et al., 2019; Macan et al., 2024).

Given the various effects plastic residues can have on soil, understanding the input sources and pathways of macro- and microplastics is essential for identifying hotspots and mitigating their accumulation. One way to identify the input sources of plastic residues is to understand the differences in their physical and chemical characteristics. Li et al. (2021) demonstrated that differences in microplastic characteristics, such as shape, colour, and polymer type, reflect different input sources. On the other hand, the occurrence of plastics with similar characteristics across distinct environments (e.g., freshwater, seawater, sediment, and soil) suggests shared pollution sources or the exchange of plastic residues between systems (Li et al., 2021; Wang et al., 2019).

One of the main sources of macroplastic inputs in agricultural soil systems is the intentional use of plastic materials in agriculture (Bläsing & Amelung, 2018; Li et al., 2022; Ren

et al., 2023). However, other agricultural management practices, such as compost application (Stefano & Pleissner, 2022), can also contribute to high macroplastic concentrations in agricultural soils. Stefano and Pleissner (2022) found 9247 items per ha^{-1} in fields treated with compost, compared to the 220 items · ha^{-1} in untreated fields, whereas Bläsing and Amelung (2018) estimated an annual input of 0.016 to 6.3 kg · ha^{-1} of visible plastic, depending on the compost application rate. In addition, littering, wind transport, and/or improper waste disposal can provide a pathway for macroplastic accumulation in soils (Bläsing & Amelung, 2018; Piehl et al., 2018; Ren et al., 2023; Stefano & Pleissner, 2022).

Microplastic input sources are more complex, as microplastics can be generated from the fragmentation of macroplastics (Ren et al., 2023). The embrittlement of agricultural plastics (Li et al., 2022), due to weathering and mechanical stress, can accelerate their fragmentation (Song et al., 2017), making them a significant source of microplastic contamination (Maqbool et al., 2024). Moreover, other main pathways for microplastic input sources in agricultural systems are tyre wear (Maqbool et al., 2023; Rehm & Fiener, 2024; Ren et al., 2023), sewage sludge, irrigation, compost, littering, and wind transport (Bläsing & Amelung, 2018; Ren et al., 2023).

Studies on microplastic contamination are widespread and important, as they provide valuable information for ecological risk assessment, determining pollution status, identifying hotspot concentrations, understanding historical trends, and assessing exposure to organisms (Shim et al., 2017). Microplastics vary considerably in terms of type and form, including different polymer types, sizes, and shapes (Shim et al., 2017; Wang et al., 2024). This variability typically limited the detection of these particles in different soil matrices, including agricultural soils (Lodh et al., 2025; Peneva et al., 2025; Wang et al., 2024). Moreover, differences in regional climate, crop type, and agricultural management will influence the abundance of microplastics (Ding et al., 2020; Haixin et al., 2022; Rillig & Lehmann, 2020; Sa'adu & Farsang, 2023). These inconsistencies are the main reason for the limited comparability between studies (Maqbool et al., 2023; Shim et al., 2017; Wang et al., 2024).

Despite the less challenging detection of macroplastics in soils (e.g., detectable to the naked eye), research on their pollution and impact on agricultural soils remains limited (Berenstein et al., 2024; Maqbool et al., 2024). Therefore, the objectives of this exploratory study are: (i) to investigate the extent of contamination by macroplastics and microplastics in different European agricultural systems where plastic has been used over the last few decades; and (ii) to analyse whether systematic patterns exist in the size distribution of plastic residues in these systems.

3.2 Materials and methods

3.2.1 Field description

To represent different agricultural systems and regions in Europe where plasticulture is intensively used, two sampling regions in Spain and Germany were chosen. In Spain, we focus on the region of Andalusia, specifically the Los Llanos and El Rocío regions, where a total of five test fields were analysed (Figure 3.1, Table 3.1). Following the Köppen-Geiger climate classification, the fields in Los Llanos and in El Rocío presented the Csa climate (temperate, dry summer, hot summer) (Beck et al., 2023). According to the FAO classification, the typical soil in the Los Llanos region (S-LL1 and S-LL2) is eutric Cambisols, eutric Regosols, and chromic Luvisols, while the El Rocío region (S-ER1, S-ER2, and S-ER3) presents humid Cambisols and dystric Gleysols (Junta de Andalucía, 2005). The mean annual temperature in El Rocío is 18ºC, the mean annual cumulative precipitation is 506 mm, and the mean annual cumulative sunshine duration is 3280 hours, while in Los Llanos the mean annual temperature is 16ºC, the mean annual precipitation is 355 mm, and the mean annual cumulative sunshine duration is 2971 hours (Deutscher Wetterdienst, 2025b).

The management practices between the two Spanish regions vary (Table 3.1, Table S1). In brief, the fields in Los Llanos presented a small-scale, less mechanised system with a history of over 15 years of mulch film use (Table 3.1, Table S1), where beans (using thin mulch film) were mainly cultivated on the analysed fields (Table 3.1, Table S1). While the fields in El Rocío presented a large-scale, more mechanised system with a history of over 20 years of mulch film use, where raspberries (using thick mulch film) were cultivated (Table 3.1, Table S1).

In Germany, we focus on an area near Nuremberg (Bavaria) and Mannheim (Baden-Württemberg), where a total of 11 test fields were analysed (Figure 3.2, Table 3.1). Both regions in Germany represent a Cfb climate (temperate, no dry season, warm summer) according to the Köppen-Geiger climate classification (Beck et al., 2023). According to FAO classification, the typical soils in the area of Mannheim are haplic Luvisols develop on loess-covered loamy to sandy river-terrace deposits (G-MA1) and calcaric Regosols developed on loess alternating with rendzic Leptosols from marlstone and limestone (G-MA2), while in the area of Nuremberg dystric Cambisols from quartzitic sandstones and conglomerates with low base status (G-NU1), cambic and haplic Podzols from sandstone and quartzite with low base status (G-NU2), and cambic and haplic Podzols from sandstone and quartzite with low base status (G-NU3 to G-NU9) (BGR, 2013) are the most important. Nuremberg has an annual mean temperature of 10ºC, a mean annual precipitation of 600 mm, and a mean annual cumulative sunshine duration of 1753 hours, while Mannheim has an annual mean temperature of 11ºC, a mean annual cumulative precipitation of 640 mm, and a mean annual cumulative sunshine duration of 1745 hours (Deutscher Wetterdienst, 2025b).

The management practices between the two German regions also vary (Table 3.1, Table S1). The test fields around Mannheim represent different crop rotations, where thin

mulch films were used regularly for approximately 10 years (Table 3.1, Table S1). The test fields around Nuremberg exhibited more distinct differences in plastic use and crops cultivated (Table S1). The fields G-NU1 and G-NU2 had a history of over 5 years of mulch film use (Table 3.1), where a variety of crops were cultivated (mainly asparagus using thick mulch films) (Table 3.1, Table S1). The fields G-NU3, G-NU4, G-NU5, G-NU7, and G-NU8 presented a different agricultural plastic use, specifically plastic film tubes, as horseradish was the only crop cultivated (Table S1). Fields where horseradish was cultivated presented a history of plastic film tube use over 4 years (Table 3.1). The other fields, G-NU6 and G-NU9, do not present any use of agricultural films (Table 3.1, Table S1).

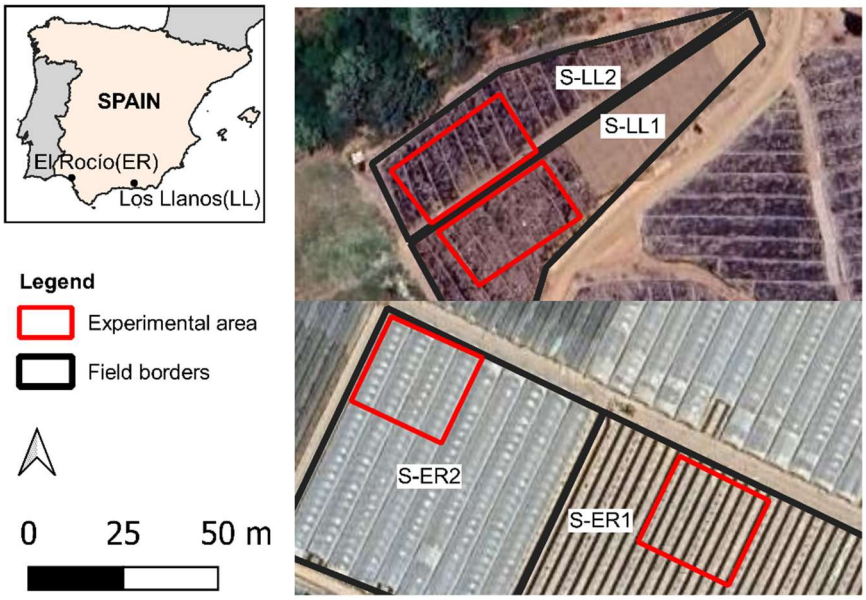

Figure 3.1: A subset of the analysed agricultural fields in Spain representing intensive use of plasticulture, where macro- and microplastic residues were analysed. For abbreviations, see Table 3.1.

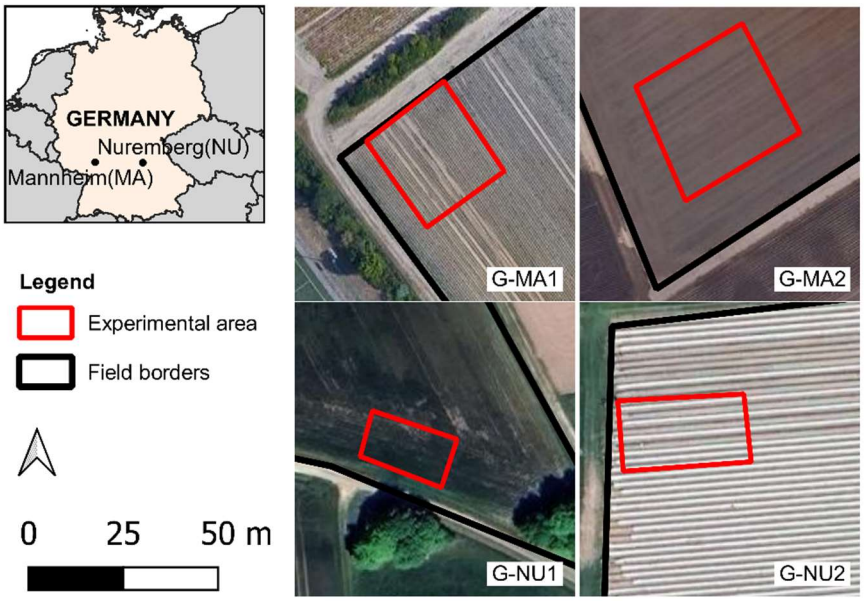

Figure 3.2: A subset of the analysed agricultural fields in Germany representing intensive use of plasticulture, where macro- and microplastic residues were analysed. For abbreviations, see Table 3.1.

Table 3.1: History of the plastic mulch film types and duration of use, as well as fertilisation with potentially macroplastic contaminated compost for the different test fields.

Filed ID‡	Region	Mulch film*	Thickness	Use of other plastics*	Plastic history	Compost*
			µm		year	
S-ER1	Spain, El Rocío	Yes	80–150	Yes	+ 20	No info
S-ER2	Spain, El Rocío	Yes	80–150	Yes	+ 20	No info
S-ER3	Spain, El Rocío	Yes	80–150	Yes	+ 20	No info
S-LL1	Spain, Los Llanos	Yes	20	Yes	+15	Yes
S-LL2	Spain, Los Llanos	Yes	20	Yes	+15	Yes
G-MA1	Germany, Mannheim	Yes	40	Yes	+11	Yes
G-MA2	Germany, Mannheim	Yes	40	Yes	+10	Yes
G-NU1	Germany, Nuremberg	Yes	125	Yes	+9	Yes
G-NU2	Germany, Nuremberg	Yes	125	No info	+5	Yes
G-NU3	Germany, Nuremberg	No	No info	Yes	+13	Yes
G-NU4	Germany, Nuremberg	No	No info	Yes	+11	Yes
G-NU5	Germany, Nuremberg	No	No info	Yes	+4	Yes
G-NU6	Germany, Nuremberg	-	-	-	-	Yes
G-NU7	Germany, Nuremberg	No	No info	Yes	+12	Yes
G-NU8	Germany, Nuremberg	No	No info	Yes	+14	Yes
G-NU9	Germany, Nuremberg	-	-	-	-	Yes

‡ S-ER1: Spain, El Rocío, no 1; S-LL1: Spain, Los Llanos, no 1; G-MA1: Germany, Mannheim, no 1; G-NU1: Germany, Nuremberg, no 1.
*See Table S1 for more detailed information.

3.2.2 Experimental design

In Spain, three fields were analysed in the El Rocío region and two fields in the Los Llanos region. In Germany, nine fields were analysed in the Nuremberg region (Bayern) and two fields were analysed in the Mannheim region (Baden-Württemberg). For most of the fields, a sampling area of 25 m x 25 m was delimited, while for some fields (five of them), this area was slightly different as the fields varied in size and shape (Figure 3.1) (see Table S1 for area overview).

For macroplastic analyses, photos of plastic residues were taken for all sampling areas (Figure 3.3). In Spanish fields, only mulch plastic residues were photographed, with a minimum

threshold size of around 5 cm. While in German fields, photographs were taken of all residue types (e.g., mulch films and littering) with a minimum threshold size of around 1 cm. The photos were taken using either a Sony Alpha 7 or a Sony Alpha 6000 camera placed on a tripod with a fixed height of 50 cm. A ruler was placed in each photo for size reference of the plastic residues (Figure 3.3).

Figure 3.3: Photos a) S-ER1, b) S-LL1, c) G-MA1, and d) G-NU2 are from mulch film residues, while e) G-MA1 and f) G-NU2 are from residues from different sources. For abbreviations, see Table 3.1.

From the 16 fields, we selected 8 fields (S-ER1, S-ER2, S-LL1, S-LL2, G-MA1, G-MA2, G-NU1, G-NU2) with a clearly documented history of mulch film use (Table 3.1) further small plastic (< 5 mm) residue analysis (Figure 3.4). In the sampling area of those fields, a composite soil sample consisting of six random samples was sampled using a soil sampler ring (Kopecky rings, Ø 5.5 cm, 4 cm height). The composite soil samples were stored in paper bags to prevent further cross-contamination with other plastic sources.

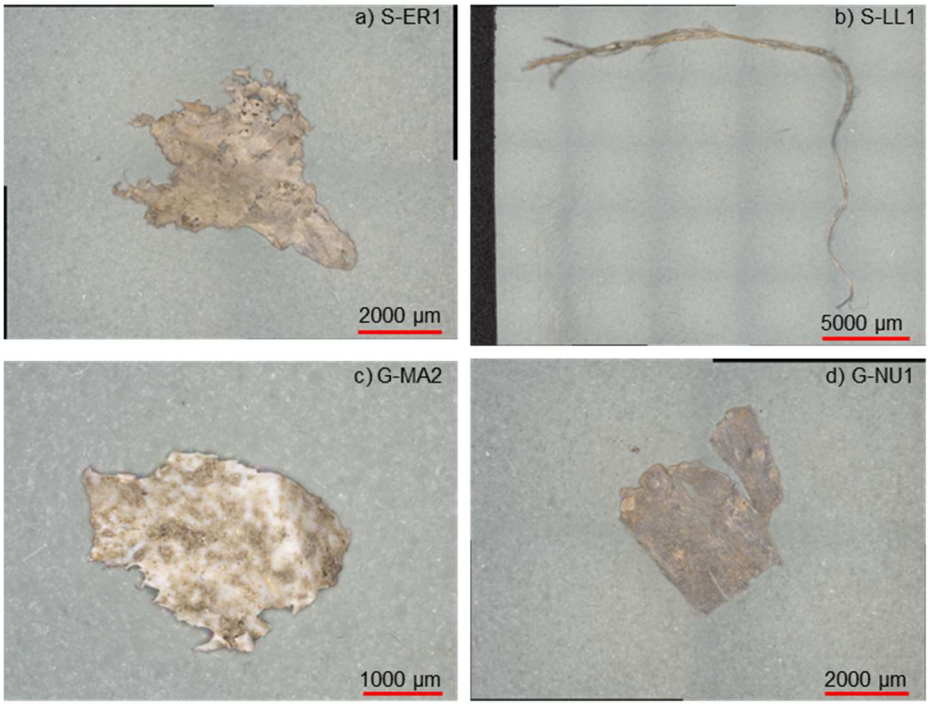

Figure 3.4: Plastic residues found on soil samples. a) S-ER1, b) S-LL1, c) G-MA2, and d) G-NU1 are plastic from unknown sources. For abbreviations, see Table 3.1.

3.2.3 Soil analysis

All collected soil samples were dried at a maximum temperature of about 30°C for two to five days to avoid any plastic loss. After drying, the composite soil sample was carefully homogenised using a mortar and pestle to prevent further fragmentation of film pieces before being sieved. A vibratory sieve shaker (Retsch AS 200 control, Retsch, Germany) was used to dry sieve the samples through 5-, 2-, and 1-mm sieves for 5 minutes at an amplitude of 1.2 mm. The soil sample in each sieve was visually inspected using a magnifying lens coupled with an LED light, and presumed plastic particles were manually collected, weighed, and stored in a plastic storage container with multiple compartments. Each compartment contained one presumed plastic residue collected from the soil.

A 3D laser scanning confocal microscope (LSM, VK-X1000, Keyence, Japan) was used to assess the surface characteristics and particle size. After the first analyses on the LSM, the particles underwent an overnight cleaning step using 0.1 M $NaP_2O_7 \cdot 10\ H_2O$ solution to remove any organic material or soil residues attached to the particles' surface. The samples were reweighted, and a second LSM analysis was performed.

The LSM is equipped with a 404 nm semiconductor laser, a complementary metal-oxide-semiconductor (CMOS) colour camera, and a depth-in-focus optical scanning system, producing height measurements, a laser reflection image, and a depth-in-focus high-resolution optical red-green-blue (RGB) image. For our analysis, samples were placed on a glass slide over a black background. Images with lens magnification of 2.5 (2.8 x 2.8 µm raster size) were taken using the coaxial and ring light together, without the semiconductor laser. The focus

variation and brightness (set to 1.4) were manually adjusted for each sample, with a height pitch of 8 µm in ultra-high-speed scanning mode. All measurement data were saved in .vk6 file format, which contains height data and RGB images.

To assess whether the analysed particles were indeed plastic residues, the particles were removed from each compartment and placed, individually, on glass slides. The glass slide containing the presumed plastic particles was subjected to a fast thermal response using a controlled hot plate (heating level 6, estimated maximum temperature of 300°C) to observe their response. The slides were placed on a hot plate for ~5 seconds. Plastic particles tend to melt when exposed to high temperatures (melting point around 115–130°C for low-density polyethylene and 130–171°C for polypropylene) (Zhang et al., 2018), while non-plastic particles, such as mineral fragments or organic matter (i.e., leaves), are not expected to change under the temperature used (loss on ignition for organic matter is around 375–800°C) (Bisutti et al., 2004).

3.2.4 Data treatment

To determine macroplastic size on the soil surface, photos of macroplastic residues (Spanish threshold: > 5 mm; German threshold: > 1 mm) were manually estimated using a ruler placed in each photo, considering the longest dimension of each residue. As mulch film should be analysed individually, and only mulch films were photographed in Spain, the photos from the German fields were also subdivided into mulch residues and other plastic residues (Figure 3.3). The main criteria for this subdivision were visual characteristics, such as colour, shape (film-like vs. 3D shape), visual surface texture, and edge characteristics. After determining the size of the macroplastic residues, we classified them into 11 size range (1–5 cm, 5–10 cm, 10–15 cm, 15–20 cm, 20–25 cm, 25–30 cm, 30–35 cm, 35–40 cm, 40–45 cm, 45–50 cm, and > 50 cm). The 1–5 cm class was a width of 4 cm, and, to be able to compare with the other classes that have a width of five cm, the particle count was normalised to a common class width of 5 cm by a factor of $^5/_4$. The other classes were not normalised. Since the area where the macroplastic residues were analysed differed (Table 3.2 and Table 3.4), after normalising the 1–5 cm class based on its width, all classes were also normalised by a common reference area of 100 m² ($\frac{residue\ quantity \times 100}{analysed\ area}$) to allow comparison among fields with different sampling areas. Their concentration on the soil surface was calculated in items per m².

For the confirmed microplastics found in the topsoil samples, their size was measured from RGB microscopy images using the Keyence software, MultiFileAnalyser (Japan), based on their longest length. The samples were also classified into 11 size ranges (0.1–0.5 cm, 0.5–1 cm, 1–1.5 cm, 1.5–2 cm, 2–2.5 cm, 2.5–3 cm, 3–3.5 cm, 3.5–4 cm, 4–4.5 cm, 4.5–5 cm, and > 5 cm). As for the macroplastic size class, the 0.1–0.5 cm class (width 0.4 cm) particle count was normalised to a common class width of five cm by a factor of $^5/_4$, and the other classes were not normalised. Since the total soil analysed for microplastic residues was different (Table

3.3), after normalising the 0.1–0.5 cm class based on its width, all classes were also normalised by a common reference weight of 0.1 kg ($\frac{residue\ quantity \times 0.1}{total\ soil\ analysed}$) to allow comparison among fields with different soil weights. Their concentration was calculated in items per kg in topsoil.

3.3 Results

3.3.1 Fields with mulch film history

Large mulch films residues on the soil surface

The number of plastic residues found in the sampling area in fields with a mulch film history is presented in Table 3.2. Even with a different threshold (Spain: mulch film residues > 5 cm; Germany: mulch film residues > 1 cm), the Spanish fields presented the highest concentration among all fields analysed, up to 0.54 items · m^{-2} (overall mean: 0.38 ± 0.15 items · m^{-2}), compared to the German fields, up to 0.02 items · m^{-2} (overall mean: 0.006 ± 0.01 items · m^{-2}) (Table 3.2). This difference in the overall mean concentration suggests the regional influence (Table 3.2). Moreover, in the case of a more mechanised farm (S-ER fields, Table S1), lower concentrations were found (regional mean: 0.28 ± 0.1 items · m^{-2}) than in a less mechanised farm (S-LL, regional mean: 0.52 ± 0.02 items · m^{-2}), suggesting the influence of management on plastic concentration (Table 3.2). In some regions (e.g., G-NU and G-MA), where fields are under the same management and climate conditions, the difference in concentrations (Table 3.2) suggests field-specific effects due to crop variety (Table S1).

After normalising the data, the field G-MA1 presented 0.16 plastic particles in the size class 5–10 cm, G-MA2 presented 0.04 plastic particles in the size class 1–5 cm (normalised class), while G-NU1 did not present any plastic residue (Table 3.2). For the other fields, the majority of the plastic residues fall into the 5–10 cm class (Figure 3.5). The size classification shows a large difference in residue quantity between the 1–5 cm and 5–10 cm classes, and a progressive decline in residues larger than 10 cm, with those larger than 25 cm becoming rare. This indicates the dominance of smaller sizes (< 10 cm) in agricultural soils. Additionally, some fields show that even within the same region (e.g., S-ER1, S-ER2, and S-ER3), where regional conditions and management practices are the same, the concentration (Table 3.2) and size class distribution (Figure 3.5) can differ, suggesting field-specific effects (management (Table S1)), highlighting the complexity of the data.

Table 3.2: Plastic residues (> 1 cm) detected in the sampling area in fields where mulch films were used.

Field ID	Area	Mulch film-based plastic residues	Plastic concentration	Regional mean concentration (± standard deviation)	Overall mean concentration (± standard deviation)
	m^2			items · m^{-2}	
S-ER1	625	125	0.2		
S-ER2	625	243	0.39	0.28 ± 0.1	
S-ER3	625	153	0.24		0.38 ± 0.15
S-LL1	630	341	0.54	0.52 ± 0.02	
S-LL2	630	322	0.51		
G-NU1	312.5	0	0	0.01 ± 0.01	
G-NU2	312.5	6	0.02		0.006 ± 0.01
G-MA1	625	1	0.002	0.002 ± 0	
G-MA2	625	1	0.002		

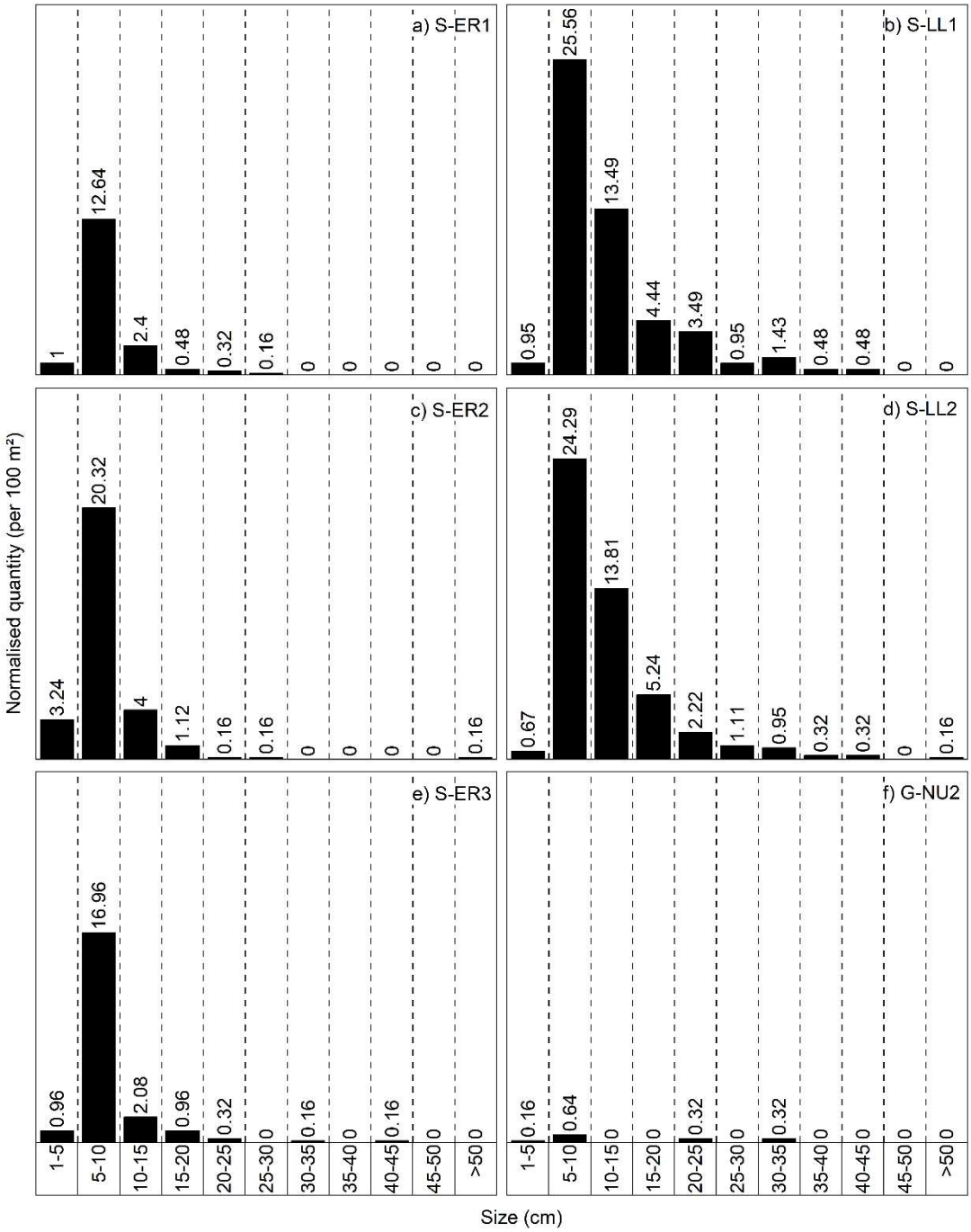

Figure 3.5: Size class of the mulch film-based plastic residue inside the sampling area in the El Rocío (a, c, and e); Los Llanos (b and d), and Nuremberg (f) regions. For abbreviations, see Table 3.1.

Small plastic residues in the agricultural soil samples

The number of plastic residues found in the topsoil with a mulch film history is presented in Table 3.3. The Spanish topsoil samples presented a higher concentration of plastic residues, up to 28 items · kg^{-1} (overall mean: 17.5 ± 8.1 items · kg^{-1}), compared to German topsoil samples, up to 19 items · kg^{-1} (overall mean: 5.75 ± 8.85 items · kg^{-1}) (Table 3.3). As for the regional and overall concentrations of plastic residues on the soil surface (Table 3.2), the differences in the regional and overall concentrations of plastic residues in the topsoil samples (Table 3.3) also suggest the regional and management influence.

Table 3.3: Plastic residues analysis in agricultural topsoil (< 4 cm depth) samples.

Field ID	Total soil analysed	Plastic residues	Plastic concentration	Regional mean concentration (± standard deviation)	Overall mean concentration (± standard deviation)
	kg			items · kg^{-1}	
S-ER1	0.790	11	14	11.5 ± 3.5	17.5 ± 8.1
S-ER2	0.973	9	9		
S-LL1	0.478	9	19	23.5 ± 6.4	
S-LL2	0.352	10	28		
G-NU1	0.630	12	19	10 ± 12.73	5.75 ± 8.85
G-NU2	0.905	1	1		
G-MA1	0.632	1	2	1.5 ± 0.7	
G-MA2	0.657	1	1		

The majority of the plastic residues fall into the 0.1–0.5 cm class (microplastic) (Figure 3.6), with residues bigger than 2 cm becoming rare. The variability between fields from the same region (e.g., G-NU1 and G-NU2) suggest the influence of differences in management (Table S1). Even though a threshold (< 5 mm) was used, bigger residues were measured due to their shape and the way their size was measured (measured considering the longest dimension).

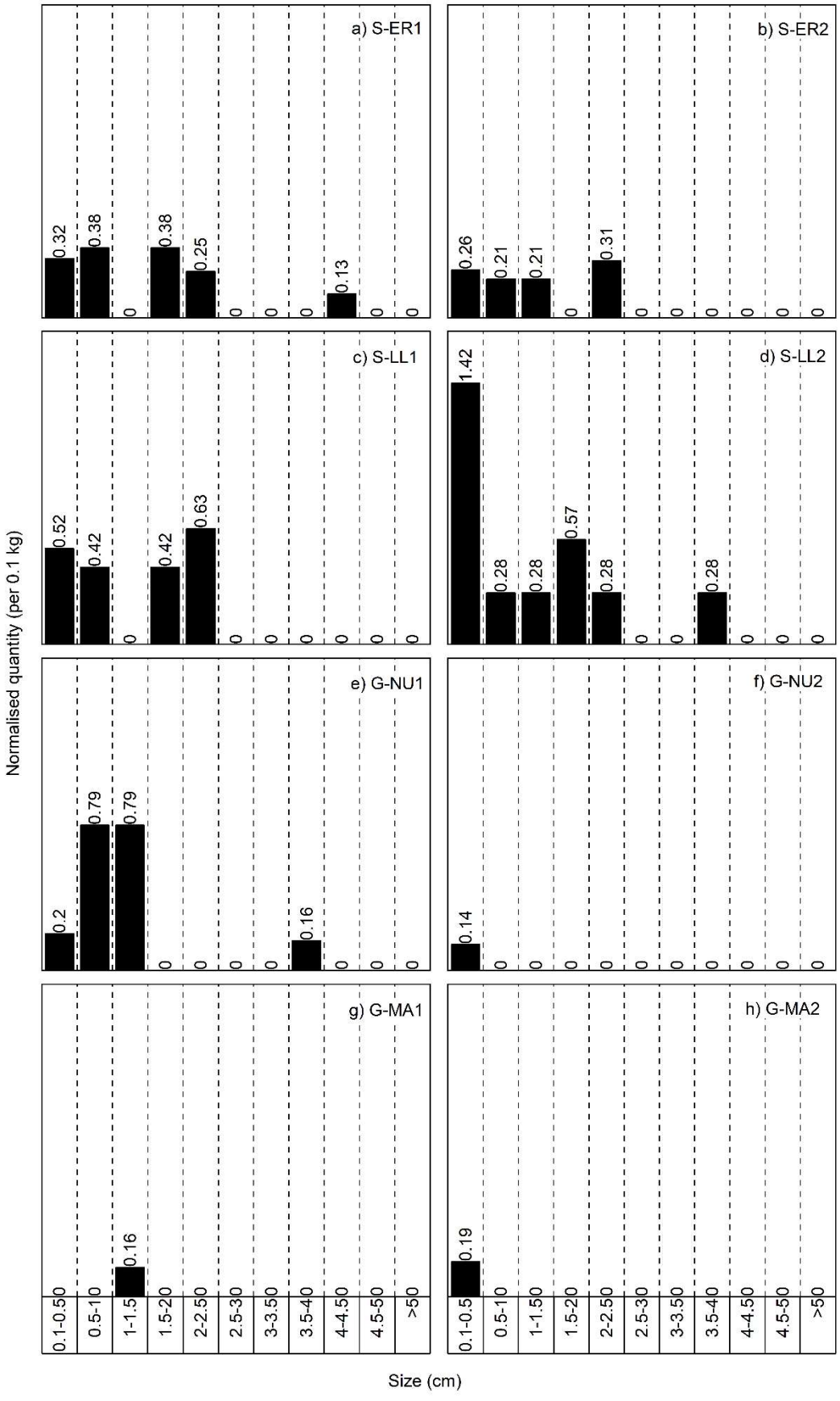

Figure 3.6: Size class of the plastic residues found in the selected soil samples. For abbreviations, see Table 3.1.

3.3.2 Fields with other agricultural plastics history

Large plastic residues on the soil surface

Besides the agricultural fields with a history of mulch film, German fields with various agricultural plastics and management histories were also analysed (Table 3.1, field description section). The number of plastic residues found in the sampling area is presented in Table 3.4. In fields where horseradish with plastic film tubes was planted and compost was applied (G-NU3, G-NU4, G-NU5, G-NU7, and G-NU8) the highest concentrations of up to 0.18 items · m^{-2} (mean: 0.05 ± 0.07 items · m^{-2}) were found, while the other fields (G-NU6 and G-NU9) were plastic residues potentially come from compost applications alone exhibit clearly lower values (mean: 0.04 ± 0.04 items · m^{-2}) (Table 3.4). It is worth noting that the origin of plastic residues from compost sources is often difficult to determine because of the variety of input sources and quality control that the compost facilities might present.

Table 3.4: Plastic residues detected in fields where no mulch films were applied. Note: The fields where horseradish was cultivated are marked in bold.

Field ID	Area	Plastic residues	Plastic concentration	Management mean concentration (± standard deviation)	Overall mean concentration (± standard deviation)
	m^2			items · m^{-2}	
G-NU3	**625**	**11**	**0.02**		
G-NU4	**1250**	**229**	**0.18**	**0.05 ± 0.07**	
G-NU5	**625**	**17**	**0.03**		
G-NU6	625	41	0.07		0.05 ± 0.06
G-NU7	**625**	**1**	**0.002**	0.04 ± 0.04	
G-NU8	**625**	**13**	**0.02**		
G-NU9	625	6	0.01		

The majority of the plastic residues fall into the class 5–10 cm, as well as G-NU7 (0.16 residues at 5–10 cm), except G-NU3 (10–15 cm class) and G-NU8 (15–20 cm class) (Figure 3.7). Figure 3.7 shows a similar pattern to Figure 3.5: a difference in residue quantity between the 1–5 cm and 5–10 cm classes, with a decline in residues larger than 10 cm, and residues larger than 15 cm becoming rare, except for G-NU8 (Figure 3.7e). As the fields share the same regional climate, it is expected that these differences are related to the less mechanised management, which increases the variability within fields (Table S1)

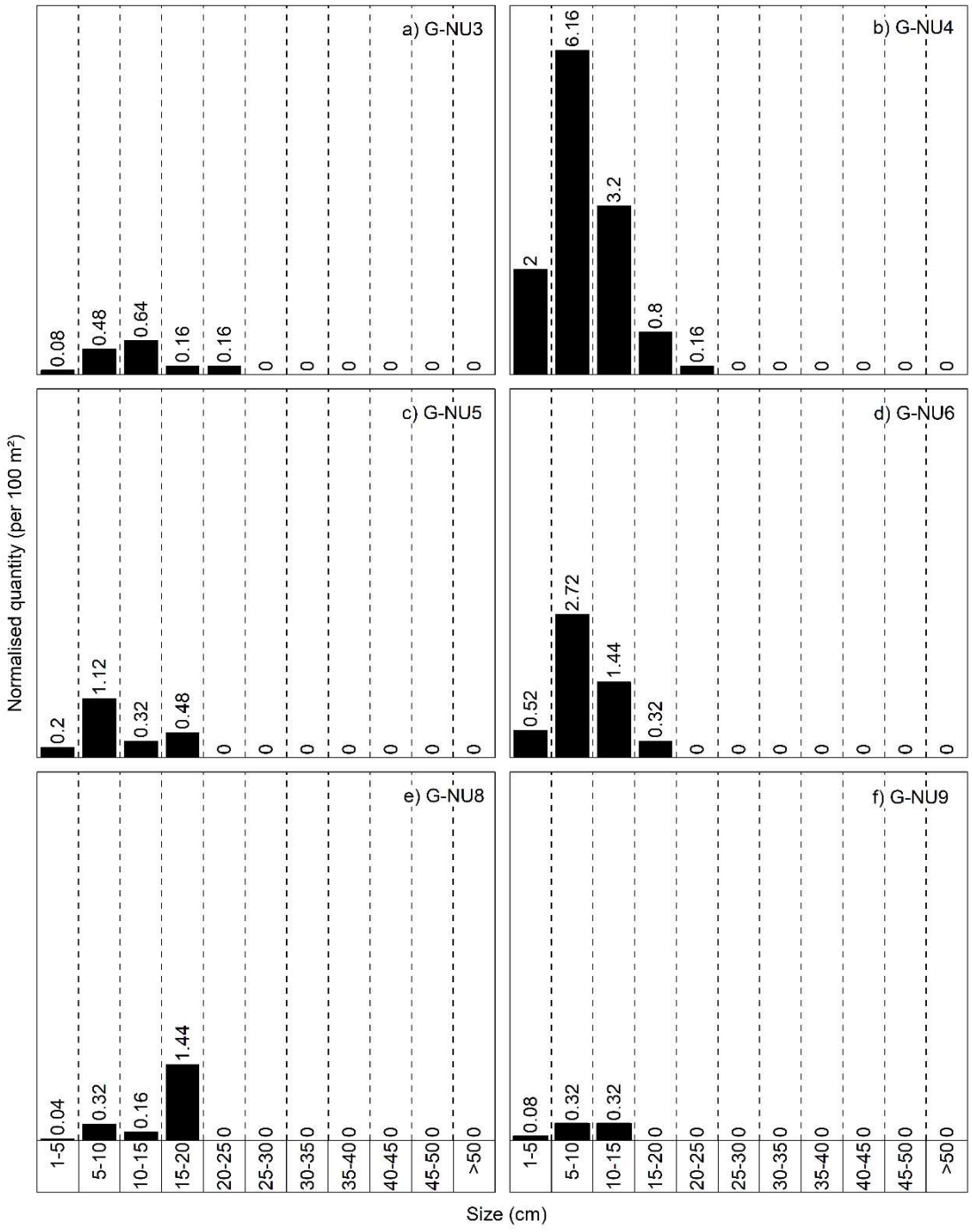

Figure 3.7: Size class of the plastic residue inside the sampling area in the Nuremberg region. For abbreviations, see Table 3.1.

3.4 Discussion

In our study, fields with different plasticulture histories, including varying cropping, soil management, plastic type use, and compost fertilisation, exhibited, as expected, large variability in concentrations of plastic residue. Regarding the macroplastic mulch film residues on the soil surface (Table 3.2), in Spain, the highest concentrations were found in the case of fields in Los Llanos region (S-LL, beans cultivation, regional mean: 0.52 ± 0.02 items · m^{-2}), where a less mechanised management was applied, with the use of mulch film (20 µm) and compost, while the concentrations in the El Rocío region (S-ER) were somewhat smaller (raspberries cultivation, regional mean: 0.28 ± 0.1 items · m^{-2}), where a more mechanised management was applied, with the use of mulch film (80–150 µm); however, there is no information about compost use (Table S1). We assume that this difference is mostly related to the management practices and mulch films characteristics (Table 3.1, Table S1). Macroplastic studies have demonstrated how management can influence their concentration (Li et al., 2022; Maqbool et al., 2024; Stefano & Pleissner, 2022; Weber et al., 2022). Different crops require the use of mulch films with different characteristics, which can be linked to distinct pollution patterns (Li et al., 2022; Steinmetz et al., 2022). Thinner agricultural plastic films are more prone to disintegration during removal (Li et al., 2022). For example, Manzano et al. (2019) reported that thinner mulch films (10 µm) present a lower average removal (25% of removal) compared to thicker mulch films (25 µm, 90% of removal). In Germany, the highest concentrations were found in the case of fields in the Nuremberg region (G-NU, mixed crops, regional mean: 0.01 ± 0.01 items · m^{-2}), where a less mechanised management was applied, with the use of mulch film (125 µm) and compost, while the concentration in the Mannheim region (G-MA) were smaller (mixed crops, regional mean: 0.002 ± 0 items · m^{-2}), where also a less mechanised management was applied, with the use of mulch film (40 µm) and compost (Table S1). However, in the case of the German fields, fields where thicker films were used (G-NU) presented higher concentrations (Table 3.2). Since both farms had less mechanised management, we assume the difference in concentrations is due to their specific management practices, as the crop varies between the fields.

For the microplastic residues in the topsoil, the German fields, presented concentrations of up to 19 items · kg^{-1} for the G-NU fields (regional mean: 10 ± 12.73 items · kg^{-1}) (mixed crop, Table S1), compared to up to 2 items · kg^{-1} for the G-MA fields (regional mean: 1.5 ± 0.71 items · kg^{-1}) (mixed crop, Table S1) (Table 3.3). While for the Spanish topsoil samples, plastic concentrations of up to 28 items · kg^{-1} were found for the S-LL fields (regional mean: 23.5 ± 6.34 items · kg^{-1}) (beans, Table S1), compared to up to 14 items · kg^{-1} for the S-ER fields (regional mean: 11.5 ± 3.53 items · kg^{-1}) (raspberry, Table S1) (Table 3.3). S-LL fields had a history of using thinner mulch films compared to the S-ER fields (Table 3.1). While G-NU fields had a history of using thicker mulch films compared to G-MA fields (Table 3.1). The findings on the German topsoil samples contradict the existing literature, where thinner mulch films will present higher concentrations of plastic residues (Li et al., 2022; Steinmetz et al., 2022; Steinmetz & Schroder, 2022; Uzamurera et al., 2023; Zhou et al., 2020). Nevertheless, fields with higher concentrations of macroplastic residues on the soil surface (Table 3.2) also

presented higher concentrations of microplastic residues in the topsoil samples (Table 3.3), suggesting a direct link between macroplastic residues and subsequent microplastic residues. Studies have shown that macroplastics in agricultural soils can increase microplastic concentrations (Berenstein et al., 2024; Li et al., 2022; Piehl et al., 2018; Weber et al., 2022). Berenstein et al. (2024) analysed and compared the concentrations of macroplastic (> 2 cm) residues in a previously studied Argentinean field (analysed for plastic concentration in 2015) that has not been used for any agricultural activity since then. They found that the macroplastic residue concentration significantly increased (2015: 13 ± 4 items · m^{-2} soil; 2022: 36 ± 11 items · m^{-2}), suggesting fragmentation (Berenstein et al., 2024) and the persistence of residues.

The German fields where horseradish was cultivated (G-NU3, G-NU4, G-NU5, G-NU7, and G-NU8) (Table S1) exhibited the highest numbers of macroplastic residues (management mean: 0.05 ± 0.07 items · m^{-2}) (Table 3.4), compared to other crop types in Germany (G-MA1, G-MA2, G-NU1, and G-NU2, Table S1) (overall mean: 0.006 ± 0.01 items · m^{-2}) (Table 3.2). Horseradish requires a very specific type of cultivation, and to the best of the author's knowledge, no literature was found to compare concentrations. To cultivate horseradish, plastic film tubes are used surrounding the horseradish root, which are introduced to restrict the spread of unwanted side roots. At harvest, the root is removed together with the plastic film tubes, but often plastic residues remain in the soil (Figure 3.8).

For the macroplastic residue size class, most of them fall into the 5–10 mm size class (Figure 3.5 and Figure 3.7) in both regions and plastic types (mulch and plastic film tubes). For the microplastic residues (Figure 3.6), the majority of the residues fall into the size class 0.1–0.5 cm. Even though the topsoil samples were sieved to 5 mm, bigger particles were found, both because of their shape (i.e., fibres) and because some particles were crumpled and passed through the sieve. Another influence on the size classification was the use of microscope images, as the longest size was used when measuring and classifying the plastic residues.

Figure 3.8: Plastic film tube residues in agricultural soil.

The literature on macroplastic concentration remains scarce (Berenstein et al., 2024), which makes comparisons with existing literature difficult. For comparison purposes, the data reported on Figure 3.9 were normalised to the same depth (< 10 cm) (lower graph) and unit (items · m^{-2}) (both graphs). Our results, presented in Figure 3.9, were also normalised using the same parameters, since the data could not be directly compared with those in Table 3.2 and Table 3.3. Nevertheless,

similar macroplastic concentrations to ours have been reported in the literature (Figure 3.9). Even though macroplastic residues can present an advantage in analytical techniques (visible to the naked eye) (Maqbool et al., 2024), the reported literature on topsoil samples of those residues varies by several orders of magnitude (Figure 3.9, lower graph), highlighting the complexity of macroplastic data. This complexity comes from the poor documentation of factors influencing field contamination over the years, including limited information on field management, plastic characteristics, duration of use, removal, compost, and so on. However, to the best of the author's knowledge, no study discusses these influences, which makes a deeper understanding and comparability among studies even more difficult.

Additionally, variation in concentration can be explained by regional influences (Haixin et al., 2022). When comparing different regions with similar plasticulture (i.e., mulch films with similar thickness (Table 3.1), Spanish fields presented higher concentrations of macroplastics (overall mean: 0.38 ± 0.15 items · m^{-2}) compared to the German fields (overall mean: 0.006 ± 0.01 items · m^{-2}) (Table 3.2). Similar results were found for the plastic residues in the topsoil, with a higher concentration in the Spanish fields (overall mean: 17 ± 8.1 items · kg^{-1}) compared to the German fields (overall mean: 5.75 ± 8.85 items · kg^{-1}). However, these regional influences are also dependent on the plastic characteristics (Haixin et al., 2022; Manzano et al., 2019), with thicker films expected to present better mechanical properties (Xiong et al., 2023).

On a global scale, Wrigley et al. (2024) reported a global mean microplastic concentration of 2900 ± 7600 items · kg^{-1} based on 89 studies (553 fields) across the globe. However, it is necessary to consider that these studies differ in terms of plastic size range, management, and methodological techniques. From the 89 studies analysed by Wrigley et al. (2024), 81% were from Asia, accounting for only 6 different countries. In general, the limited number of studies worldwide masks the real-world scenarios of microplastic pollution (Wrigley et al., 2024). Additionally, microplastics can interact with soil aggregates, hampering their detection (Rehm et al., 2021; Zhang & Liu, 2018), with studies showing 72% of plastic particles associated with soil aggregates (Zhang & Liu, 2018).

Incomplete documentation, limited characterisation and sampling methodologies, different regional influences, management practices, input sources, differences in size classification, shape, polymer type, and reporting units hinder the detection and comparability of plastic residues across studies (Bläsing & Amelung, 2018; Hartmann et al., 2019; Peneva et al., 2025; Wang et al., 2024; Yu & Flury, 2021). To build a clearer understanding of plastic risks in agricultural soil, more standardised protocols and information about sampling and extraction procedures, reporting and documentation, considering climate, management practices, and historical data, are urgently needed, as this will reduce variability between studies, allowing careful comparison (Miller et al., 2021). Overall, considering all the information discussed above (i.e., the variability in region, different management practices, and poorly documented information) helps to explain the wide variation in data reported in this and across other studies.

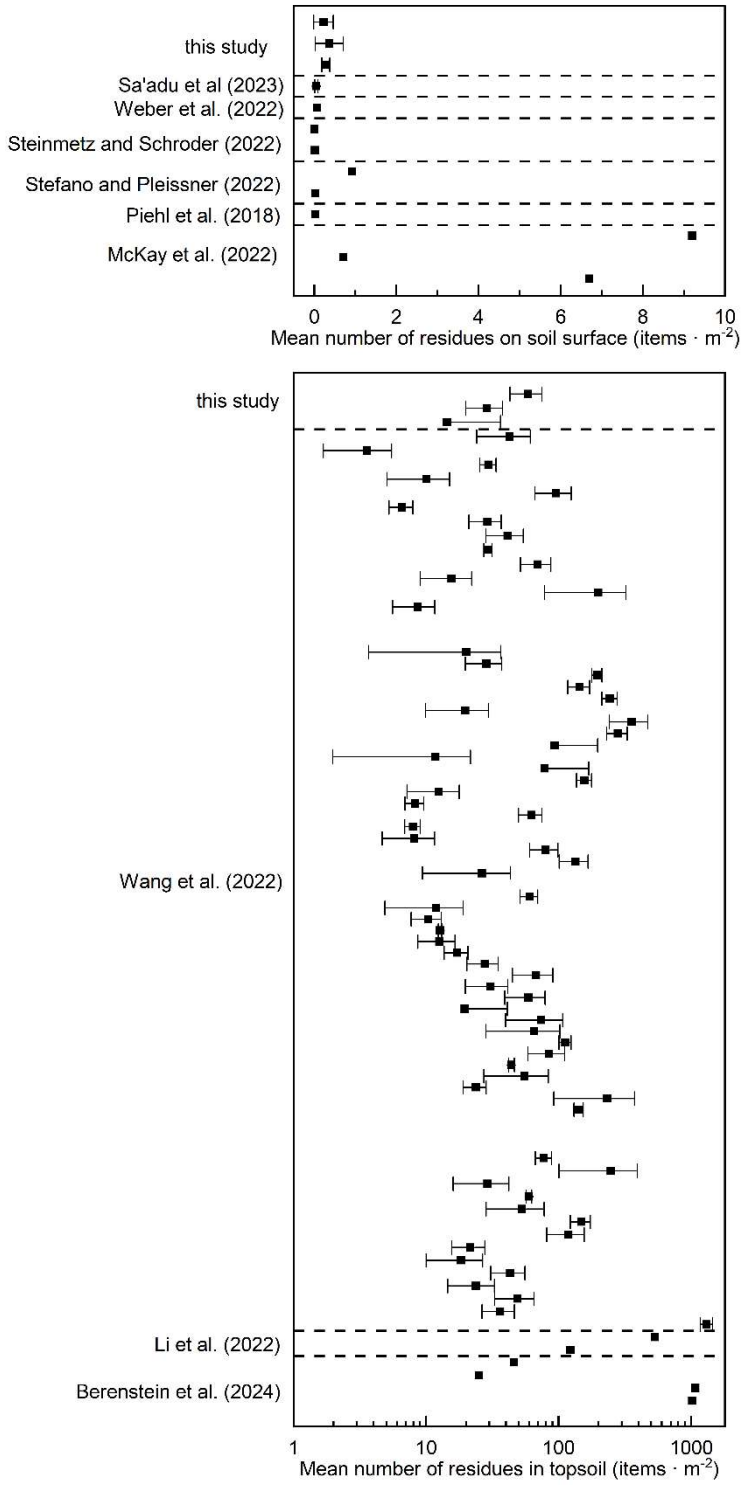

Figure 3.9: Comparison of macroplastic concentration in this study with the literature. The upper graph compares macroplastic concentrations at the soil surface. The lower graph compares (in log scale) macroplastic concentrations in the topsoil. Note: for comparison, all values were adjusted to items · m^{-2} and the topsoil to a depth of < 10 cm.

3.5 Conclusion

Our study highlighted the influence of regional climates and management practices on macro- and microplastic concentrations in agricultural fields. Although macroplastics are easier to detect, studies focusing on them are still limited; nevertheless, our results are consistent with the literature. Moreover, limited studies that consider other influences, such as climate and management, hinder comparability across regions, a comprehensive understanding of plastic contamination in agricultural soils, and the evaluation of environmental implications at regional and global scales. Additionally, poor documentation of agricultural plastic management over the years (types of agricultural film used, use duration, removal, compost, and so on) hampers a holistic understanding of plastic contamination in agricultural soils, as the sources of plastic contamination can vary. Despite these limitations, our dataset helped reinforce that mulch film alone cannot explain plastic concentration variation, highlighting the roles of diverse factors (e.g., management practices, input sources, and regional characteristics). More standardised protocols could help account for these differences, clearly distinguishing their influence on plastic concentration and enabling the creation of mitigation strategies.

Biodegradable mulch films affecting surface runoff, soil erosion, and plastic loss from farmland[2]

4

4.1 Introduction

Plastics are used in agriculture in large quantities (i.e., 15.6×10^9 kg globally in 2021) (Plastics Europe, 2022), a practice often called plasticulture. Mulch films make up a large share of agricultural plastics, and are used to regulate soil temperature, improve water use efficiency, suppress weeds, and also in soil solarisation (Gao et al., 2021; Hann et al., 2021; Steinmetz et al., 2016). Single mulch films can be used for one or multiple cropping seasons, provided that they are thick enough to remain intact and resist fragmentation. However, thin mulch films (< 25 µm thickness) are known to undergo substantial weathering and readily fragment, releasing plastic particles (Ghimire et al., 2020; Ren et al., 2021; Zhang et al., 2021). These include macroplastics (> 5 mm) and microplastics (< 5 mm), both of which can continue to undergo fragmentation. For conventional mulch films composed of stable polymers such as low-density polyethylene (LDPE), a non-biodegradable polymer (Zumstein et al., 2016), released residues persist in soils and lead to plastic accumulation. Conversely, biodegradable plastic mulch films should achieve 90% mineralisation to CO_2 in a soil environment within an incubation period of two years in the laboratory (EN 17033, 2018). These films are made of biopolymers, such as starch, and synthetic polymers, such as bio-based polylactic acid (PLA) or fossil-based poly (butylene-adipate-co-terephthalate) (PBAT) (Gao et al., 2021).

When applied to soils repeatedly, the concentration of plastic residues from mulch films in soils may increase over time when composed of non-biodegradable conventional polymers (Li et al., 2022). Depending on the size of the plastic residues and their concentrations, these film residues can alter the physical characteristics of soils, e.g. reducing pore connectivity and affecting soil structure and water movement (Gao et al., 2021). Plastic residues in soils have also been shown to impact soil bulk density, porosity, and saturated hydraulic conductivity (Y. Qi et al., 2020). These changes in soil physical properties, especially the reduced pore connectivity and infiltration capacity, potentially affect surface runoff generation and soil erosion.

The existing literature on the effects of mulch films on surface runoff generation and soil erosion mainly focuses on measurements during the application period of the mulch films (with intact films on the soil). For example, Zhang et al. (2013) reported a substantial increase in runoff and linear erosion in agricultural fields using mulch film strips, specifically in the exposed soil surface between the strips and during early plant-growth stages. Others have

[2] An adapted version of this chapter is under review in Soil and Tillage Research as: Cugler Moreira, A. C., Wille, F., Batista, P.V.G., Sander, M., Wilken, F., Fiener, P. (2026). *Biodegradable mulch films affecting surface runoff, soil erosion, and film fragment loss from farmland.*

reported that the areas covered by mulch films are protected from soil detachment by raindrop impact and sheet flow, hence reducing overall soil erosion (Zhang et al., 2014). However, the trade-offs between the soil protection afforded by mulch films and the increased erosion on exposed soil surfaces between film strips are unclear. It is noteworthy that these effects of mulch films on surface runoff and soil erosion during early crop-growth stages should occur regardless of their type (conventional vs. biodegradable), as both film types are expected to be structurally intact at the beginning of the growing season.

Differences in soil surface runoff and erosion between conventional and biodegradable mulch films might occur after crop harvest. At this stage, conventional films are mainly removed, while the not-yet-biodegraded part of the biodegradable films is incorporated into soils by tillage. The latter practice makes biodegradable mulch films economically attractive, as it reduces labour and disposal costs (Sander et al., 2023). Yet, studies have found biodegradable mulch film residues on test fields after four years of being applied (Ghimire et al., 2020), suggesting that the actual time needed for complete biodegradation of mulch film residues in soil under in situ conditions is longer than in laboratory certification tests. Biodegradability in the field is expected to depend on film characteristics, soil properties, and the local climatic conditions (Sander et al., 2023). Consequently, these biodegradable mulch film residues might affect surface runoff and soil erosion for some years after being applied.

Changes in surface runoff and erosion caused by the effects of residual plastic mulch films in soil might also facilitate the transport of macro- or microplastic residues to adjacent ecosystems. However, relatively little research has focused on the lateral transport of plastic residues from agricultural soils, especially after biodegradable mulch films are tilled into the soil under representative field conditions. Some information exists for conventional plastics. For instance, Zhang et al. (2022) added different sizes/shapes of polyethylene terephthalate and polyethylene (PE) microplastics in a soil-leaching experiment under natural and simulated rainfall conditions. This study reported a rainfall intensity-dependent migration of microplastics in both lateral and vertical directions, with smaller particles (< 1 mm) showing higher mobility compared to bigger particles (1–5 mm). Fibres and films showed higher mobility compared to particles and foams (Zhang et al., 2022). Similar results were reported by Han et al. (2022); smaller microplastic particles (< 1 mm) and higher rainfall intensity (60 mm) increased microplastic mobility, due to surface runoff. These authors also found that vegetation can lower the lateral plastic transport by 20% (Han et al., 2022).

Rehm et al. (2021) used a series of rainfall simulations to measure high-density polyethylene (HDPE) microplastic transport from arable soils. They found a mean enrichment ratio of plastic in the delivered sediment compared to the plastic in topsoil of 3.95 ± 3.71 (mean ± standard deviation) of coarse microplastic (250–300 µm) and 3.17 ± 2.58 of fine microplastic (53–100 µm) particles. However, none of the above studies assessed the potential effects on soil hydrological and sedimentological behaviour caused by biodegradable mulch films being tilled into the soil after harvest, and how this influences the lateral transport of film residues by surface runoff and soil erosion.

This study aims to analyse the effects of biodegradable mulch films ploughed into topsoil after regular use for one growing season on surface runoff, soil erosion, and the loss of components of the biodegradable film following heavy rainfall. To this end, we performed rainfall simulation experiments on six different plots. In three plots, biodegradable mulch films were exposed on the soil surface for six months and tilled into the soil shortly before the rainfall simulations. Three other plots without mulch film were used as controls.

The following hypotheses were tested: (i) Incorporating biodegradable mulch films into topsoils by tillage at the end of the vegetation period leads to a decrease in infiltration capacity and, hence, an increase in surface runoff and soil erosion in case of heavy rainfall events. (ii) Shortly after tillage, most film residues still consist of macroplastic fragments. Hence, most film residues are not transported with runoff and erosion. (iii) However, the microplastic fraction in the soil will be preferentially transported due to the lower density compared to soil particles.

4.2 Materials and methods

4.2.1 Site description and experimental design

Our rainfall experiment was performed at the Erosion and Runoff Laboratory (EARL) experimental site in Eastern Bavaria (48°29’28’’N; 13°17’53’’E). The experimental site represented an area of about 6 ha on a south-facing slope with a gradient between 8 and 11% (Bavarian State Research Center for Agriculture, 2024). The area had a long history of intensive agricultural use but no history of plasticulture. The site has an annual mean temperature of 8.3°C and a mean annual precipitation of 957 mm (Deutscher Wetterdienst, 2025a). Using data from 1961 to 2020, the Köppen Climate classification on the experimental site region is Cfb (temperate, no dry season, warm summer) (Beck et al., 2023).

At the upslope, northern part of the experimental site, six plots (1.6 m x 4.5 m; area 7.2 m²) were installed for this study on a homogeneous slope with a gradient of 11%. The plots were bordered by metal sheets reaching 0.1 m deep into the soil and a collector funnel as an outlet (Figure 4.1). The plots had a minimal distance of 2 m from each other. The plots with the biodegradable mulch film and the controls were set up in March 2023, six months before the rainfall simulations. Treated and control plots were randomly arranged. The texture of the plots’ topsoil (depth < 0.15 m) was silt loam (15% sand, 58% silt, and 27% clay) with a topsoil organic matter content of 11.3 g C · kg^{-1}. For the experiments, we used Barbier BIONOV B, a commercially

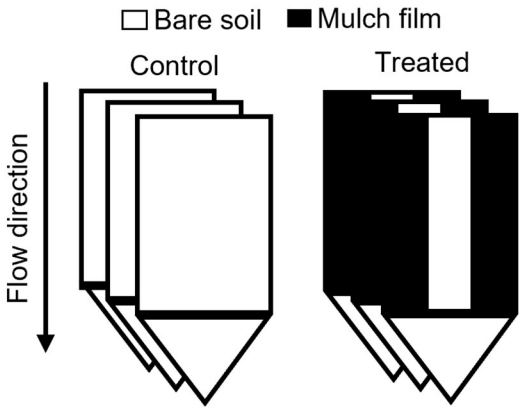

Figure 4.1: Schematic set-up of the experimental design of the triplicated plots 6 months before the rainfall simulation. The bare soil was covered with a geotextile, which was removed prior to the rainfall simulations. Mulch films were incorporated into the soil by tillage before the rainfall simulations.

available biodegradable mulch film composed mainly of PBAT and PLA polymers from NOVAMONT Mater-Bi, with a thickness of 15 µm. A commercially available polypropylene (PP) water-permeable geotextile (80 g · m^{-2}) was also used for weed and erosion control.

The control plots, without biodegradable mulch film (Figure 4.1), were covered with the geotextile (without contacting the soil surface) to prevent weed growth and erosion between plot preparation and rainfall simulation 6 months later. The mulch film plots were covered with two long pieces of biodegradable mulch film (0.73 m x 4.70 m; total film area 6.9 m²) parallel to the slope direction, representing a freshly prepared plasticulture field (Figure 4.1). The exposure for 6 months was intended to weather the mulch films as realistically as possible under natural conditions. To stabilise the films in the soil for six months, 0.1 m at each edge was buried in the soil (total area of film exposure to sunlight was 4.77 m^2). Analogously to the control plot, the geotextile (0.53 m x 4.50 m) was placed between the biodegradable film pieces, covering the exposed soil with surface contact to avoid weed growth and erosion. After 6 months, before the rainfall experiment, the geotextile was removed to prevent any influence on the transport of the biodegradable mulch film residues.

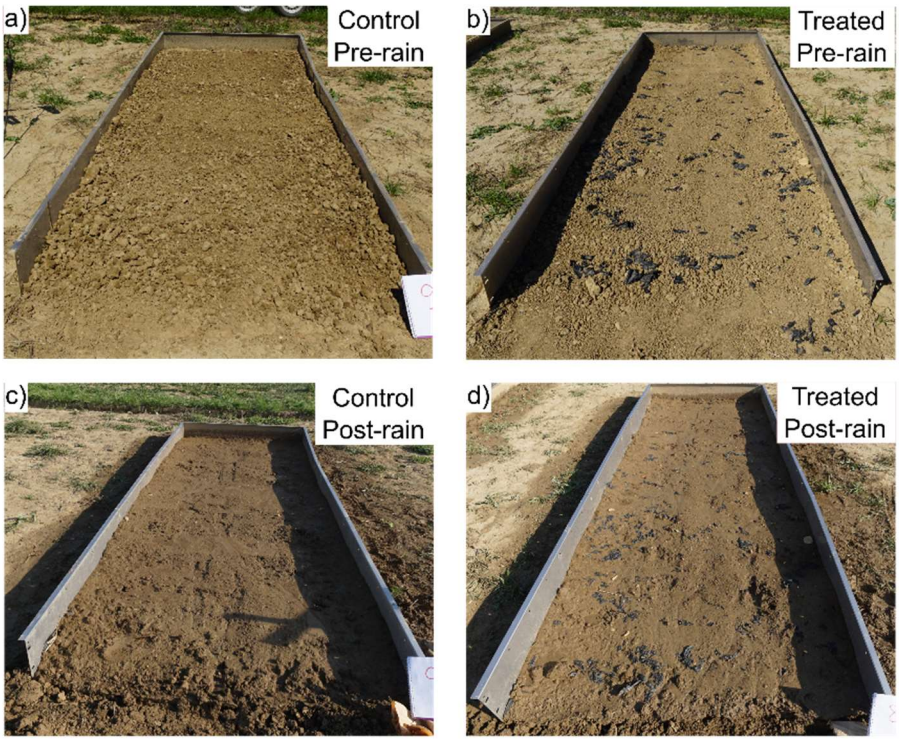

Figure 4.2: Pictures of a representative pair of the different plot treatments. The top row represents one of the triplicate plots before the rainfall simulation (pre-rain), while the bottom row represents one of the triplicate plots after the rainfall simulation (post-rain). a) and c) are the control plots; b) and d) are the plots treated with biodegradable mulch film after it had been on the soil for six months and had been tilled into the soil.

4.2.2 Rainfall simulation and sampling during experiments

Our rainfall simulations were carried out over 2 days from 11/09/2023 to 12/09/2023. There was no natural precipitation during this period, and the simulations were carried out in random order. Before the rainfall simulations, the biodegradable mulch films were first broken down into approximately 10 cm x 10 cm macroplastic residues with a spade. After this, all six plots were tilled to a 0.1 m depth with an electric garden hoe (Hecht 745; Hecht; Germany), incorporating the biodegradable mulch film into the soil, followed by compaction with a 57 kg lawn roller (GRW E 57, Gardol) (Figure 4.2a,b).

The rainfall simulation (Figure 4.3) used four swivelling nozzles (Veejet 80/100) producing a median drop diameter of 1.9 mm, a mean (± standard deviation) kinetic energy of 19.1 ± 2.3 J · m^{-2} · mm^{-1} rain and a mean drop falling velocity of 6.8 ± 0.82 m · s^{-1} to simulate an intense rain event (60 mm · h^{-1}) (Rehm et al., 2021). Before our experiment, the spatial variability of the simulated rainfall on the plot area was evaluated using 96 cups placed in a 0.3 m x 0.3 m grid, indicating a near-constant and spatially uniform rain intensity of 60.7 ± 0.15 mm · h^{-1}.

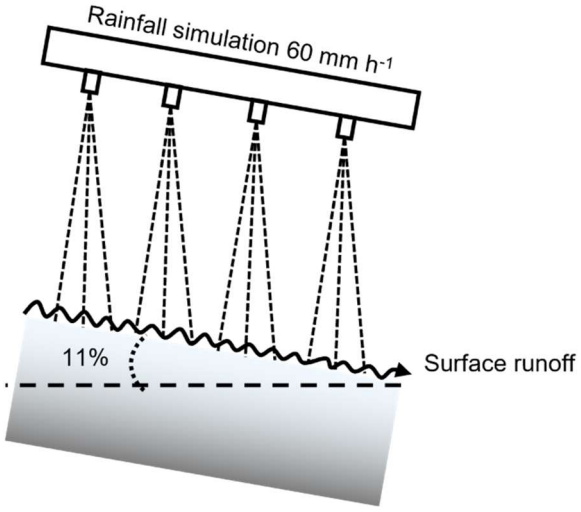

Figure 4.3: Schematic side view of the rainfall simulation.

The simulation consisted of an initial 30-minute rainfall (“dry run”), followed by a second 30-minute rainfall (“wet run”), 30 minutes after the end of the dry run. Before the dry run and after the wet run, composite soil samples (0–0.1 m) between 2 and 3 kg were taken from each plot at 10 random locations. For the dry run, about 30 seconds after the first runoff reached the downslope end of the plot, surface runoff samples were collected every two minutes via a covered stainless-steel funnel outlet (Figure 4.1), either for 90 seconds or when the 2-litre glass jar reached its capacity. As the topsoil was close to saturation at the beginning of the wet runs, the runoff samples were collected 2 minutes after the start of the rainfall experiment, with each sample jar filled in a shorter interval compared to the sampling in the dry run.

Using Equation 4.1, the runoff volume (RV) (mL) and sediment delivery (SD) (g) (see sample treatment section for further information) were gravimetrically calculated using an analytical balance with 0.01 g precision (Kern KB, Kern, Germany). JF and JE are the mass of the full and empty 2-litre glass jar (g), respectively, and water density (ρ) was considered 1 g · mL^{-1}. The sampling duration (T) (s) was recorded and used to calculate the runoff (RR) (L · s^{-1}) and sediment delivery (SR) (g · s^{-1}) rate, using Equations 4.2 and 4.3, respectively.

$$R_V = \frac{J_F - J_E - S_D}{\rho} \quad (4.1)$$

$$R_R = \frac{R_V}{T} \quad (4.2)$$

$$S_R = \frac{S_D}{T} \quad (4.3)$$

4.2.3 Sample treatment

All samples were transported to the laboratory in Augsburg (Germany) within 48 hours after the experiment started and stored at 3ºC for 20 days before further treatment. Then, all sediment samples were vacuum-filtered using an N820 vacuum pump (KNF, Germany) and a 15 cm Ø glass microfiber filter without binder with a retention rate of 1.6 µm and MGA grade (Sartorius, Germany) with a known weight. Every second sediment sample retained in the glass microfiber filter was frozen at -20ºC, together with its filter, for further analysis for biodegradable polymers content; the remaining samples were oven-dried at 105ºC for at least 24 hours before being weighed to calculate sediment delivery and runoff rate (Equations 4.2 and 4.3, respectively).

The 10 mixed composite soil samples taken before and after the rainfall simulations on each plot were analysed for biodegradable polymer content and used to calculate the enrichment/depletion ratio of biodegradable mulch film from the eroded sediments. The composite soil samples were oven-dried at 30ºC for four to seven days to ensure complete drying while maintaining the temperature sufficiently low to avoid abiotic biodegradable mulch film components hydrolysis during drying. After drying, each composite soil sample was carefully homogenised using a mortar and pestle to prevent further fragmentation of film residues before sieving the samples using a 2 mm sieve. Sample constituents larger than 2 mm, including large biodegradable mulch film residues and stones, were separated. The mulch film residues > 2 mm were analysed for biodegradable polymer contents. Subsequently, a representative subsample of 100 g was taken from each composite soil sample (2~3 kg) (< 2 mm) using an automated sampler divider (PT100, DR100, Retsch) for further biodegradable mulch film polymer analysis. All sub-samples and the remaining composite sample material were frozen at -20ºC.

The frozen filtered sediment samples and the soil composite subsamples were freeze-dried for 48 hours using either an Alpha 2-4 or Alpha 2-4 LD plus (Christ, Germany) under a 0.05 and 0.011 mbar vacuum, respectively. After the freeze-drying step, the composite soil subsamples and the sediment samples were gently milled to crush aggregates that had formed during freeze-drying. Each composite subsample and each sediment sample (together with their respective glass microfiber filter, to avoid any possible losses due to retention on the filter pores) were put in a cellulose extraction thimble (VWR, 516−0252 or Machery-Nagel, 645025). Subsequently, the composite and sediment, plus the filter, samples were weighed.

4.2.4 Biodegradable polymer solvent extractions

The polymer quantification methodology is based on Cerri et al. (2025) and Nelson et al. (2020). In brief, the composite or sediment samples in the cellulose thimbles were solvent extracted (9:1 (v/v) vol% mixture of chloroform and methanol) to dissolve and extract both PBAT and PLA from the samples. Two different extractors were used: a manual Soxhlet extractor in which dried sample volumes were less than ~20 g (heating bank: R306S, Behr Labor Technik; extractor chambers: EZ30H, Behr Labor Technik), and an automated solvent extractor that allowed for larger sample volumes and was therefore used for samples containing more than ~20 g (extractor: Universal Extractor E-800, Büchi, Germany). The thimbles were capped with either the glass microfiber filter for the sediment samples or a PP mesh for the composite soil samples and then placed in the extraction chambers. First, a methanol pre-extraction (HPLC grade, Merck) (30 min on the manual extractor; 10 extraction cycles on the automated extractor) was performed to remove background extractable soil and sediment organic matter that could interfere with polymer quantification. Next, PBAT and PLA were extracted from sediment and composite soil using a 9:1 (v/v) vol% mixture of chloroform and methanol (both HPLC grade, Merck) (60 minutes on the manual extractor; 10 cycles on the automated extractor). The solvent containing the extracted polyesters (PBAT and PLA) was evaporated until a few millilitres remained. The extract was then left to dry at room temperature overnight.

Mulch film residues (> 2 mm) collected from the composite soil samples were directly dissolved in 50–70 mL chloroform. Given the high concentrations of PBAT and PLA in these samples, 5 mL aliquots were subsequently taken and dried overnight, prior to preparation for quantitative proton nuclear magnetic resonance spectroscopy (^{1}H NMR) analysis.

4.2.5 Biodegradable polymer quantification by ^{1}H NMR

All dried sample extracts and aliquots were reconstituted in deuterated chloroform (99.8 atom% for NMR, Thermo) containing an internal standard 1,4-dimethoxybenzene (DMB, > 99% purity, Acros Organics) at 2 mg · mL^{-1}. Before q-^{1}H NMR analysis, reconstituted samples were centrifuged for 10 minutes at 13,300 rpm (Micro Star 17, VWR, USA) to separate any impurities (i.e., black particles (presumably carbon black as photo-stabiliser) and other chloroform-insoluble fillers that were present in the mulch film), and 1 mL was subsequently used for q-^{1}H NMR analysis.

The ^{1}H NMR spectra were acquired with a Bruker Avance III 400 MHz NMR spectrometer equipped with a Bruker 5 mm BBFO 400 MHz Z-Gradient probe, and the following acquisition parameters are described in Cerri et al. (2025). Spectra were processed in Mnova 14.3.3 as follows: spectra were first referenced to the peak of non-deuterated chloroform at the chemical shift of 7.26 ppm, then phases and baselines were manually corrected, and lastly the selected polymer peaks were manually integrated: i.e., the peaks at chemical shifts characteristic of the terephthalate aromatic protons of PBAT at ~8.10 ppm (T), the methylene protons in the butylene terephthalate unit of PBAT at ~4.40 ppm (BT), the

methylene protons in the butylene adipate unit of PBAT at ~4.10 ppm (BA), as well as PLA at ~5.16 ppm, and DMB at ~6.84 ppm (for spectra illustration see Figure S4). The amount of extracted polymer was calculated according to the following equations:

$$m_x = \frac{M_{w_x} \cdot a_x}{H_x} \cdot \frac{n_{DMB} \cdot H_{DMB}}{a_{DMB}} \quad (4.4)$$

$$n_{DMB} = \frac{c \times V}{M_{DMB}} \quad (4.5)$$

$$m_{PBAT} = m_{BA} + m_{BT} \quad (4.6)$$

$$T\text{-}content = 100\% \cdot \frac{a_{BT}}{a_{BA} + a_{BT}} \quad (4.7)$$

Where x refers to either T, BT, BA, or PLA, mx to the respective mass (mg), Mwx to the molecular weight of the repeat unit or internal standard (i.e., BA = 200.06 mg · mmol^{-1}, BT = 220.08 mg · mmol^{-1}, PLA = 72.06 mg · mmol^{-1}, and DMB = 138.17 mg · mmol^{-1}), ax to the area of the selected peak (arbitrary unit), Hx to the number of protons represented by the chosen peak (T and BT = 4 protons, BA = 4 protons, PLA = 1 proton), nDMB to the number of DMB moles (mmol), HDMB the number of protons of the DMB aromatic group (4 protons), c to the concentration of the internal standard solution (mg DMB · mL^{-1} deuterated chloroform), and V to the volume (mL) of the standard solution used reconstitute each sample.

PBAT mass was calculated by summing BA and BT (Equation 4.6). We note that BT can be quantified by both the T peak at a chemical shift of ~8.10 ppm and the BT peak at ~4.40 ppm; however, at low amounts of mulch film, the integral of the T-peak was often larger than the integral of the BT peak. We ascribe this to overlap with soil organic matter signals at ~8.10 ppm and used the BT peak at ~4.40 ppm for quantification. Furthermore, at low PBAT amounts, the BA peak at ~4.10 ppm was often overlapped by peak contributions from co-extracted soil organic matter and could therefore not be accurately integrated without risking over-quantification of the BA-repeat unit. In these cases, only the BT amount was calculated. Lastly, since PLA is present at only ~3 wt% in the pristine mulch film, amounts were often too low to produce a peak that could be integrated. In this case, only PBAT or BT masses could be calculated. When possible, the molar ratio of terephthalic acid to total diacid of PBAT (T-content, mol%) was calculated by dividing the area of the BT peak by the sum of the areas of the BA and BT peaks (Equation 4.7).

Selected extracts from the methanol pre-extractions underwent reconstitution and ^{1}H NMR analysis to detect if any PBAT or PLA were extracted during this step, which served to remove co-extractable soil organic matter. Neither PBAT nor PLA were detected in the methanol pre-extracts. PE (both high- and low-density) is not chloroform soluble and thus does not interfere with the PBAT and PLA quantification. Therefore, HDPE bags and flasks were used to store samples when necessary.

4.2.6 Data treatment

Runoff did not start at the same time after the beginning of the rainfall simulations for every plot. Therefore, the sediment samples were not always taken at the same time points for all replicate plots. In the case of the mulch film treatment, we calculated the estimated average and standard deviation of the biodegradable polymer concentrations for a given time point using the replicate samples from the three independent plots that were closest in time. For example, for the sample collected after four minutes of rainfall, a mean was calculated by considering values from minutes two, four, and six for all three replicates. A typical mean and standard deviation were calculated for the replicate control plots where the runoff samples were taken simultaneously.

The measured runoff and sediment data were linearly interpolated between measurements to derive a continuous hydro-sedigraph and calculate the total runoff and sediment delivery for each rainfall simulation in each plot. For the dry run, because of the higher infiltration capacity, the time 0 was set when runoff first reached the downslope end of the plot, 30 seconds before collecting the first sample. As the soil was already saturated for the wet run, the time 0 was set when the simulation started, which was 2 minutes before collecting the first runoff sample. The mean and standard deviation of the total amount of runoff and soil losses were calculated for each treatment. The runoff coefficient (C) (arbitrary unit) was calculated using Equation 4.8.

$$C = \frac{Q}{P \times A} \tag{4.8}$$

Where Q is the overall runoff volume from each rainfall run on each plot (L), P is the overall rainfall volume applied during the rainfall simulation ($L \cdot m^{-2}$ or mm), and A is the plot area (m^2). Using the OriginPro 2023b software (OriginLab, USA), we performed a Kruskal-Wallis test followed by Dunn's test to compare the differences between treatments for the runoff coefficient, the cumulative sediment, polymer total concentration, and enrichment data. The OriginPro 2023b software (OriginLab, USA) was also used to produce all the figures.

The enrichment or depletion ratio (ER) of biodegradable mulch film in the delivered sediments compared to the biodegradable mulch film concentration in the topsoil was calculated by averaging the enrichment ratio of each replicate (Equation 4.9).

$$ER/DR = \frac{mean\ biodegradable\ polymer\ concentation\ in\ sediment}{mean\ biodegradable\ polymer\ concentration\ in\ topsoil} \tag{4.9}$$

An ER > 1 indicates an enrichment of biodegradable plastic in delivered sediments, while an ER < 1 indicates a depletion compared to topsoil samples. Note, we separately calculated the biodegradable mulch film concentration in the topsoil using the biodegradable residues in the fine soil fraction only (< 2 mm) and the fine soil fraction plus the biodegradable mulch film residues > 2 mm. For both size fractions, an ER is calculated separately to assess potential enrichment of the different size fractions.

4.3 Results

4.3.1 Surface runoff and soil erosion

The plots where biodegradable mulch film was incorporated displayed higher runoff mean rates for dry and wet runs (Figure 4.4a and b) and higher sediment delivery means rates for the dry run (Figure 4.4c), but not the wet run (Figure 4.4d). Moreover, runoff on the wet run from the plots containing the worked-in biodegradable mulch presented a higher runoff compared to the control plots, indicating a reduction in the infiltration capacity of the soils treated with the biodegradable mulch films (Figure 4.4a).

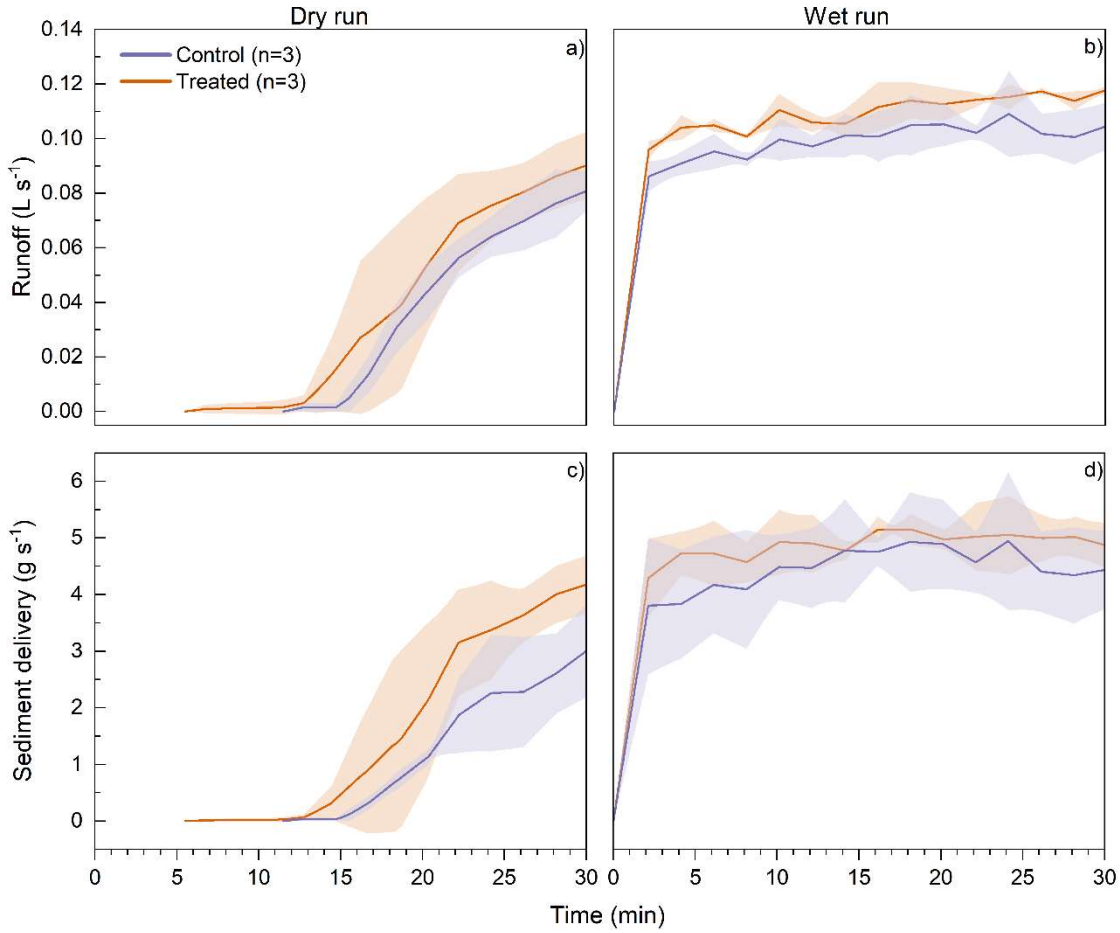

Figure 4.4: Runoff (a and b) and sediment delivery (c and d) rates from the dry and wet runs. Treated represents the plots where the soil included tilled-in biodegradable mulch film pieces. The shaded area represents the standard deviation of each treatment (n=3).

Overall, the mean cumulative sediment delivery and runoff coefficients (Figure 4.5) mirrored the differences in sediment delivery and runoff rates (Figure 4.4). As expected, the wet runs resulted in higher runoff and sediment delivery values for both treatments than the dry runs. Between treatments, there was a statistical difference ($p < 0.05$) for the runoff coefficient, but only for the wet run. There was no statistical difference in cumulative sediment delivery between the treatments in both runs Figure 4.5).

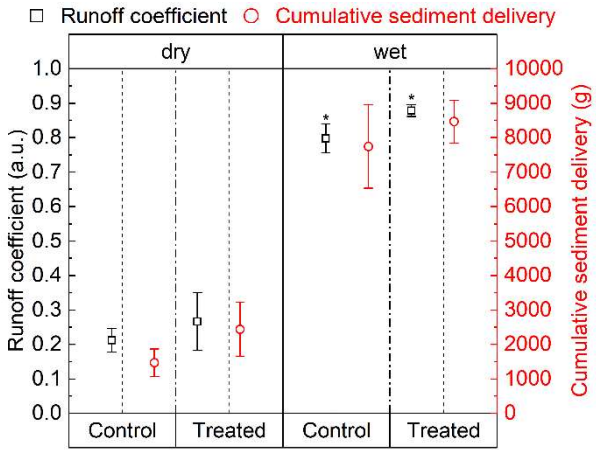

Figure 4.5: Mean runoff coefficients and cumulative sediment delivery for the dry and wet runs from different treatments. Error bars indicate the standard deviations (n=3) of each treatment. Treated represents the plots treated with biodegradable mulch. "*" represents the treatments that are statistically different ($p < 0.05$) from each other.

4.3.2 Biodegradable polymer in soil and sediment samples

The amounts of biodegradable polyesters (PBAT (in terms of both BT and BA), and PLA) extracted by the solvent extraction method and quantified by ^{1}H NMR analysis are shown in Table 4.1. The amount of biodegradable polyester found on the control plots (Table 4.1) was small, as only one of the three replicates presented absorbance in the BT chemical shift range. The origin of this absorbance is unclear, which can be either from the background noise of the soil organic matter or cross-contamination between plots. Since the individual polyester components (BA, BT, PLA) were often present at very low levels and sometimes not detectable by the ^{1}H NMR technique, and because this study focuses on the total mass of transported biodegradable mulch film, we use a sum of the biodegradable polyester in the following (Figure 4.6).

Table 4.1: Biodegradable mean polymer concentration values from different treatments. "-" represents data not collected.

Treatment	Sample	Rainfall simulation run	Biodegradable polymer (µg g^{-1}) (± stdv)			
			BT	BA	PLA	Total polymer
Mulch film	Topsoil < 2 mm	Dry and wet	2.6 (±0.72)	0	0	2.6 (±0.72)
Control			0.13 (±0.23)	0	0	0.13 (±0.23)
Mulch film	Topsoil > 2 mm	Dry and wet	27.24 (±15.13)	26.45 (±14.8)	2.81 (±1.55)	56.51 (±31.49)
Control			-	-	-	-
Mulch film	Sediment	Dry	1.6 (±0.83)	0.41 (±0.69)	0.41 (±0.37)	2.41 (±1.65)
Control			-	-	-	-
Mulch film	Sediment	Wet	2.05 (±1.1)	1.30 (±0.97)	0.41 (±0.44)	3.76 (±2.5)
Control			-	-	-	-

For PBAT, the T-content was determined (Equation 4.7), given that biodegradation of PBAT has been observed to increase the T-content. The topsoil > 2 mm sample of the plot treated with biodegradable mulch film presented a T-content of 48.5 (± 0.2) mol%. In contrast, the PBAT in the pristine mulch film had a T-content of 47 mol%, indicating that the field-aged

mulch film may have undergone some biodegradation before it was incorporated into soil. The BA unit of the biodegradable mulch film is more susceptible to biodegradation than the other units (i.e., BT and T), therefore, increasing the T-content.

During the rainfall simulations, the runoff samples of the plots treated with biodegradable mulch film displayed a low biodegradable polyester concentration during the dry run, as the amount of soil eroded was very small (Figure 4.6). However, particularly after 14 minutes of simulated rainfall in the dry run, a slight increase in biodegradable polymer concentration which matched the dynamics in runoff and sediment delivery rates, suggesting that the polymer was transported with sediment (Figure 4.4a and c). During the wet runs, the plastic concentration in the delivered sediments was more variable than in the dry run, with some peaks in polymer concentrations in specific time intervals, e.g. 2–6 min, 14–18 min, and 26–30 min. However, the high variability represented by the standard deviation (Figure 4.6) indicates that probably only one of the replicates presented a higher concentration of biodegradable polymer. This higher concentration could be connected to random pieces, most likely larger pieces, being transported with the surface runoff.

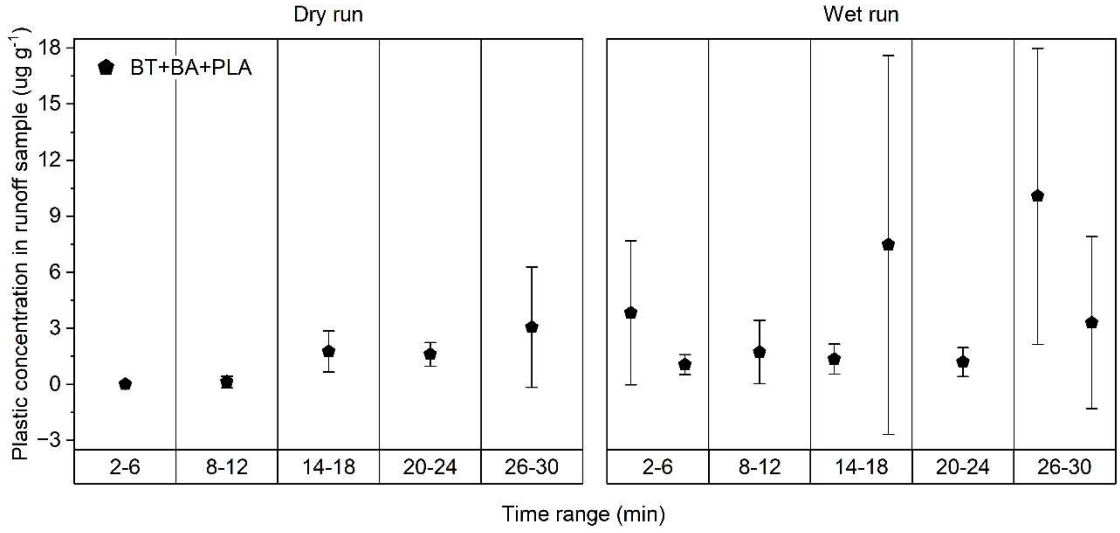

Figure 4.6: Butylene terephthalate unit of PBAT (BT), butylene adipate unit of PBAT (BA), and polylactic acid (PLA) mean concentration (n=3) in sediment samples from the plot treated with biodegradable mulch film, 6-month exposed. Error bars represent the standard deviation (n=3).

The ER (Equation 4.9) for the < 2 mm fraction (sediment samples divided by the fine composite soil samples) was highly variable for the less dense, fine polymer fraction (Figure 4.7), and encompassed unity for both the dry and wet runs. Hence, neither enrichment nor depletion of biodegradable polymers can be established. For the < and > 2 mm fraction (sediment samples (< 2 mm) divided by the composite soil samples (< 2 mm) plus larger biodegradable film residues found in the plot's topsoil (> 2 mm)), there was a depletion in biodegradable polymer concentrations in eroded sediments (ER < 1, Figure 4.7), which indicated that the runoff did not transport larger mulch film residues.

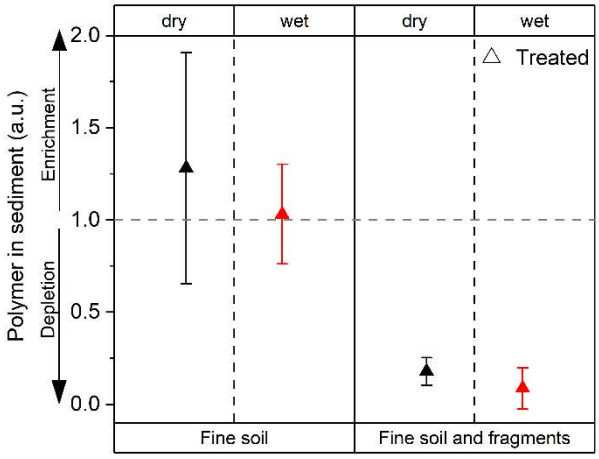

Figure 4.7: Enrichment/depletion ratios of biodegradable polymer in the delivered sediment relative to (i) fine soil (< 2 mm) and (ii) fine soil and biodegradable residues (> 2 mm). The error bars represent the standard deviation calculated from the mean across three different plots. Preferential polymer transport is demonstrated by an enrichment factor > 1, while depletion is demonstrated by a factor <1.

4.4 Discussion

Not surprisingly, six months after establishment on the soil surface, a substantial amount of the biodegradable mulch film was left and was tilled into the topsoil. As expected from the small mass of the thin biodegradable film residues mixed into 10 cm of topsoil, the overall concentrations of large (> 2 mm) and small (< 2 mm) plastic residues within the topsoil were small. However, as long as macroplastic residues are left in topsoils after ploughing, infiltration rates may be reduced (Jiang et al., 2017; Y. Qi et al., 2020). Hence, there is a higher probability of increased surface runoff in the case of heavy rainfall events.

This effect is shown for the surface runoff dynamics, especially for the wet run (Figure 4.4). The influence of biodegradable residues on the infiltration rates is also evident from the higher runoff coefficients of the treated over the control plots (mean difference for the dry and the wet run is 25.5% and 10%, respectively (Figure 4.5)). However, only for the wet run, the difference in runoff coefficient between the treated and control plots was significant ($p < 0.05$) (Figure 4.5). The missing significance on the runoff coefficient in case of the dry run, resulting from a larger variability in runoff, is probably associated with differences in surface moisture at the beginning of the dry runs, an effect intentionally levelled out while performing wet runs. While the agronomic implications of buried mulching films lie outside the scope of this study, the observed hydrological alterations, specifically higher runoff in the wet run linked to lower soil-water infiltration, suggest that the drought resilience of agroecosystems may be affected, as less precipitation water is retained in the soil. While it is expected that with an increase in surface runoff, soil erosion and sediment delivery would also increase on the plots with the ploughed-in biodegradable film, leading to a higher runoff coefficient, especially in the wet run,

and cumulative sediment delivery (Figure 4.5), no statistical difference was found for the sediment delivery in both runs between treatments.

In general, any effects on surface runoff and sediment delivery are expected to fade out with ongoing fragmentation and biodegradation of the larger biodegradable mulch film residues. Based on laboratory incubation tests, a prerequisite for the certification of biodegradable mulch films is that they have to achieve 90% mineralisation within two years (EN 17033, 2018). However, field studies indicate that biodegradation is slower under real-world soil conditions compared to laboratory incubations. For example, Ghimire et al. (2020) reported that about 34% of the material remained in the soil four years after the incorporation of biodegradable films. Similar results were reported by Sintim et al. (2020), who found that 27% of the biodegradable mulch film remained after 36 months in soil. Moreover, it is important to note that we tested a 1-time mulch film application, while, especially in vegetable production, it is common to use and plough in biodegradable mulch films in consecutive years. Under such conditions, higher concentrations of mulch film residues can be expected even in the case of a 90% biodegradation within two years. Generally, it might be assumed that an increase in erosion also leads to an effective loss of biodegradable film residues. However, in our study, we could not demonstrate any significant effects. Nevertheless, our data support hypothesis 2 that film residues > 2 mm are rarely transported via sheet and small rill erosion processes, such as those in our experiment. Hence, residues > 2 mm are strongly depleted (more than a factor of 4) in the delivered sediments in our experiment (Figure 4.7). Overall, it is expected that depletion would be less pronounced when upscaling from plots to catchments, where more unselective rill and ephemeral-gully erosion processes come into play (Schiettecatte et al., 2008). Sources of uncertainty should be considered when interpreting the data. Measurements included plot replicates; however, no replicate samples were taken within the plots, underestimating the analytical variability. Variability in runoff start times could also have an effect on treatment comparisons. Additionally, low biodegradable polymer content concentrations often prevented peak integration, possibly underestimating the total plastic transported. Overall, these factors suggest that the uncertainties in plastic residue transport can be higher than indicated and should be interpreted cautiously.

The smaller film residues (< 2 mm) behave more or less like bulk soil with a slight enrichment in the case of the dry runs (not significantly different from an ER = 1; Figure 4.7), so hypothesis 3 cannot be verified. Density could be one of the reasons the enrichment of the lighter biodegradable plastic particles does not follow studies with conventional microplastics, which all indicate an enrichment due to their lower density compared to the soil particles (Rehm et al., 2021), as microplastics made of PE present a lower density (~0,9 g cm^{-3}) compared to the biodegradable mulch film used in this study (~1.2 g cm^{-3}). The material shape studied could have also played a role (Zhang et al., 2022). While Rehm et al. (2021) used spherical particles, this study used flat films. It could also be that most of the residues in the < 2 mm fraction are still too large to be transported (Zhang et al., 2022), or the mulch film residues are more strongly interacting with soil particle surfaces as compared to PE microplastic particles previously used.

Given the size of the biodegradable mulch film residues in the topsoil, the potential of transport in erosion-prone areas into adjacent ecosystems, such as in aquatic systems, will be reduced (Figure 4.7), where these biodegradable polymers that were designed for biodegradation in soils under aerobic conditions may persist (Nauendorf et al., 2016; Shi et al., 2024; van Grinsven & Schubert, 2023; Wei et al., 2021). However, simultaneously with a decreasing effect on surface runoff and potentially erosion due to the ongoing fragmentation of the larger film residues (see above), it can be expected that more small residues will emerge, leading to a decrease in the depletion of mulch film residues in the delivered sediments.

4.5 Conclusion

A rainfall simulation experiment was carried out to gain insight into the influence of biodegradable mulch film pieces on soil surface runoff, erosion, and biodegradable particle transport. The freshly ploughed-in mulch film significantly increased surface runoff coefficients compared to the control, but only in the wet run. The lack of statistical significance for the dry run reflects the larger variability in soil moisture between the triplicate plots. The sediment delivery didn't present a significant difference between treatments.

Together with soil, biodegradable film residues were eroded from the plots, despite a one-time application of the biodegradable mulch film. Still, the total amount was small (2.41–3.76 µg per g of soil). In our experiment, most film residues in the topsoil were relatively large (partly a few centimetres) after tilling films pre-exposed for 6 months. As these large residues are hardly transported with sheet erosion, their overall residue concentration in the delivered sediments was substantially smaller compared to the topsoil, reducing the film residue transport potential in case of heavy rainfall events shortly after ploughing in the mulch film.

However, it is important to note that our experiment only represents the condition shortly after the mulch film was ploughed in. As time progresses and residues biodegrade, it can be assumed that the reinforcing effect of the larger film residues on surface runoff and erosion will fade out. Associated with this process, the depletion of mulch residues in the delivered sediment will also decrease with time. This means that as the mulch film in the topsoil further fragments as part of the biodegradation process, the small film residues are expected to show increased transport through soil erosion and, therefore, according to their residual concentration in the topsoil.

General discussion 5

5.1 Thesis objectives

To achieve the overarching aim of this thesis, field and laboratory experiments were conducted. The hypothesis that (i) the interaction between ultraviolet radiation and mechanical abrasion would increase degradation and microplastic formation was proven. As was (ii) the influence of climate and management practices on plastic accumulation. In general, Chapters 2 and 3 demonstrate that a single dominant process does not govern plastic degradation and accumulation; instead, a combination of factors, influenced by material properties, environmental conditions, and management practices, governs it.

These findings show us that the degradation behaviour and the factors influencing the accumulation of plastic waste in different agricultural systems are dynamic. Furthermore, the interactions among these factors highlight the potential environmental consequences of plastic residues in agricultural soils, which require further study. As evidenced in Chapter 4, there was (iii) transport of plastic waste to surrounding ecosystems and influence on hydrological and erosion processes.

In general, these results show that the life cycle of agricultural plastics affects not only the environment in which they are used, but also the surrounding environment. The complexity of the degradation and fragmentation processes, coupled with the environmental impacts of agricultural plastics presented in this thesis, alerts us to possible consequences that need further study.

5.2 Degradation processes and plastic accumulation in agricultural soil

Agricultural plastics were subjected to relevant environmental stressors present in the agricultural environment (photodegradation and mechanical abrasion). The experimental findings (Chapter 2) highlight how exposure to ultraviolet radiation can promote degradation, while abrasion can accelerate it by removing degraded surface layers and exposing the inner parts of the polymer. Although microplastic formation was not directly quantified, this thesis still provides a plausible explanation for plastic fragmentation and accumulation in agricultural soils. Several studies, using similar weathering conditions, that quantified microplastic formation, reported the same cyclical process (degradation and removal of degraded layers) (Bhattacharjee et al., 2023; Ouyang et al., 2023; Song et al., 2017; Yang et al., 2022).

Although only a few relevant stressors were considered in this thesis, under real environmental conditions, the rate of this degradation cycle and the potential for fragmentation are highly variable (Chamas et al., 2020). This will depend on plastic-specific characteristics (e.g., additive content or thickness) (Haixin et al., 2022; Niu et al., 2025), ultraviolet exposure, temperature, humidity (Fairbrother et al., 2019), soil particle movement (Song et al., 2017),

and management practices (Li et al., 2022), among other influences (Ren & Ni, 2022; Xu et al., 2025; Yu et al., 2021).

Field observations on accumulation and spatial distribution revealed large variability in macro- and microplastic concentrations across different agricultural soils (Chapter 3). However, similar results were reported in the literature (Figure 3.9) (Braun et al., 2023; Harms et al., 2021). Furthermore, the results suggest that macroplastics can serve as a source of microplastics over time (Berenstein et al., 2024; Li et al., 2022; Weber et al., 2022).

Different regional conditions and management practices, such as compost, sewage sludge use (Brandes et al., 2021), and tillage (Maqbool et al., 2024) can influence the concentration and distribution of plastic residues in agricultural soil, indicating that not only the degradation processes influence the abundance of plastic residues (Bläsing & Amelung, 2018; Song et al., 2017) in soil, but rather a combination of multiple factors (Haixin et al., 2022; Wrigley et al., 2024) (Chapter 3). Multiple input sources and poor documentation of plastic use also contribute to variability in concentration. Overall, this thesis highlights that plastic pollution in agricultural soils is context-dependent, and understanding the complexity of factors that contribute to it remains necessary.

Given the variability of factors that affect degradation and accumulation, finding a single condition or variable that explains these mechanisms would be unwise. Understanding the integration of different environmental factors is essential to understanding the large accumulation of plastic in the environment and its effects. Studies should put into perspective different combinations of factors that affect the life cycle of plastic, since, once in the environment, it is almost impossible to control how these residues behave or how they will be affected by the environment.

5.3 Methodological approaches

The challenges of comparing plastic residue concentrations across studies, due to methodological variability, also need to be considered (Wang et al., 2024) (Figure 3.9). This thesis shows that, in addition to environmental factors and the characteristics of the materials used, equipment limitations increase variability, hindering a deep understanding of the subject. Additionally, differences in sampling and extraction methods, plastic size classification, and units reported continue to hinder comparability, requiring methodological standardisation (Hartmann et al., 2019; Peneva et al., 2025; Wang et al., 2024; Yu & Flury, 2021).

Although macroplastics are easier to detect (Maqbool et al., 2024), they are rare, whereas microplastics can be more abundant but harder to detect due to detection limits (Peneva et al., 2025; Wang et al., 2024). As shown in this thesis, spatial distribution can increase data variability. Studies have shown how management practices can influence spatial variability (Maqbool et al., 2024; Weber et al., 2022). This demonstrates how the sampling method will influence how realistic and representative the data will be (Yu & Flury, 2021).

Although standardisation can restrict scientific freedom (Hartmann et al., 2019), its absence limits the capacity to make quantitative comparisons or mass balances of plastic inputs and persistence in soils (Wrigley et al., 2024). Even when similar management regimes were applied, specific conditions, such as regional climate, led to variability in the data (Chapter 3), highlighting, once again, that plastic residue accumulation in soils is a complex, heterogeneous process. These variabilities highlight the challenges and limitations in detecting and quantifying plastic residues across systems, and generalised assumptions should be avoided.

Standard methodologies can shape research and influence future mitigation strategies (Hartmann et al., 2019). However, a standard methodology for such complex materials and matrices is a dream that could take a few years to be developed, if it will ever be. Before achieving standard protocols, new, more sensitive techniques are required to detect smaller plastic residues and fully understand their gravity and real effects on the environment. Additionally, proper documentation of plastic use and a detailed report that accounts for as much variability as possible should be the main focus, as they will increase understanding and comparability in the long term. Until then, studies on plastic pollution must be interpreted within the context of plastic characteristics, management practices, and climate conditions, reinforcing the context-dependence of plastic accumulation.

5.4 Effect of mulch film residues on surface runoff and soil erosion

The results from the rainfall simulation experiments (Chapter 4) showed that incorporated soil-biodegradable mulch films can influence surface runoff and soil erosion. Previous reports showed that mulch film residues, regardless of their origin, affect soil properties and are transported to surrounding ecosystems (Han et al., 2022; Jiang et al., 2017; Y. Qi et al., 2020; Rehm et al., 2021). However, these effects can be linked to the size, shape (Zhang et al., 2022) or concentration of plastic residues (Rehm et al., 2021).

This thesis shows how the transport of residues is size-dependent. Small residues (< 2 mm) tend to be enriched in sediment delivery and transported to surrounding ecosystems, whereas larger residues (> 2 mm) are not easily transported (Chapter 4). Repeated use of soil-biodegradable materials can contribute to the accumulation of their residues, either because field biodegradation rates vary between different environmental conditions (Ghimire et al., 2020; Sintim et al., 2020) or because 10% is allowed to persist (EN 17033, 2018). While further fragmentation increases the transport of small residues, the effect on hydrological and erosion processes should decrease. Additionally, when certified soil-biodegradation residues are transported to surrounding ecosystems, e.g., aquatic environments, their biodegradability certification is lost (SAPEA, 2020), becoming persistent pollutants (van Grinsven & Schubert, 2023).

Although soil-biodegradable films are an alternative to conventional plastics to address plastic pollution (Gao et al., 2021), their environmental impacts still require further study. As

demonstrated in this thesis, environmental conditions influence the degradation (Chapter 2) and the distribution of plastic residues (Chapter 3). Thus, understanding the biodegradation of these films under different conditions is necessary to evaluate the true benefits of their use.

5.5 Biodegradable or conventional mulch films?

Agricultural plastics are an essential piece in modern agriculture, and this scenario is unlikely to change soon. With increased awareness of plastic pollution, attempts were made to reduce it in agricultural soils. The trade-off between conventional mulch films and soil-biodegradable alternatives is often promoted as a sustainable alternative (Gao et al., 2021), as soil-biodegradable films can biodegrade in the soil (Dentzman & Goldberger, 2020). However, the findings of this thesis demonstrate that they can still have environmental impacts (Chapter 4).

It is known that the biodegradability of soil-biodegradable mulch films is influenced by different environmental conditions (e.g., temperature and humidity) (Sander et al., 2023; Sintim et al., 2020). Since biodegradability can vary with different environmental factors, it is very important to understand how these factors influence the biodegradation of soil-biodegradable plastics. Once their biodegradability is compromised, they are expected to behave and affect soil ecosystems like conventional mulch films (SAPEA, 2020).

As an example of potential soil-biodegradable accumulation, simulated scenarios in which soil-biodegradable films are used every year and their biodegradation follows either a linear or an exponential trend over different time periods were calculated (Figure 5.1). Assuming that every year 15 g · m^{-2} of soil-biodegradable mulch film is applied, and their biodegradability behaves as expected, 90% mineralisation in two years (EN 17033, 2018), if a linear trend is assumed, around 9 g · m^{-2} would still be in the soil after two years of mulch film use (the 10% remains of the first year application plus the remains after one year of the second year application) (Figure 5.1). If the linear trend is still assumed, the accumulation of residues would reach 35 g · m^{-2} in five years (Figure 5.1).

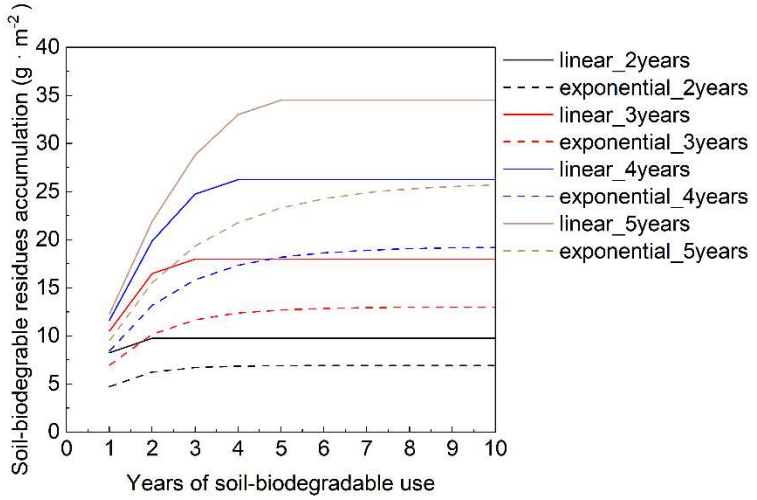

Figure 5.1: Yearly accumulation of soil-biodegradable residues, considering linear and exponential biodegradation over the years.

Although these hypothetical values consider the perfect scenario, where 90% of the residues are mineralised (EN 17033, 2018), and disconsidering variabilities or influences on biodegradation (Sander et

al., 2023; Sintim et al., 2020), their accumulation would be higher in locations where conventional mulch film is heavily used (Huang et al., 2020). Huang et al. (2020) found an average macroplastic (> 5 mm) concentration of 8.36 g · m^{-2} across 394 sampling fields in China, where fields have used conventional mulch films for over 30 years. Therefore, the trade-off between soil-biodegradable and conventional mulch films should not be seen as a universal solution to plastic pollution, and their use should be considered in light of the environmental conditions and management practices.

Thus, more studies that consider different conditions (e.g., temperature, humidity, and soil texture) should be conducted to better understand the behaviour of soil-biodegradable mulch films across different environments. Furthermore, because soil-biodegradable residues affect erosion and hydrological processes (Chapter 4), a possible alternative to controlling plastic pollution and reducing its impacts on soil and the environment would be to remove soil-biodegradable mulch films after use rather than incorporating them into the soil.

5.6 Complementary environmental solutions

The findings of this thesis suggest that mitigating agricultural plastic pollution through soil-biodegradable mulch films is insufficient. Alternatives, such as investing in and improving the recycling strategies of conventional films, may offer additional advantages (Agriculture Plastics Environment, 2021). Recycling may reduce plastic inputs more effectively than repeated incorporation of soil-biodegradable materials (Figure 5.1) that require specific environmental conditions (EIP-AGRI Focus Group, 2021).

At the same time, reducing overall plastic use and reconsidering the necessity of agricultural plastics in certain cropping systems can be a long-term strategy. However, this strategy could influence productivity and resource efficiency. Avoiding agricultural plastic altogether would be impractical, as it would require substantial changes to agricultural practices and would likely limit food production.

Preventing agricultural plastic residues from entering agricultural soil and the environment in general should be the priority. Better to avoid than to remediate. However, to achieve this goal, new and better plastic materials should be developed that will both avoid plastic pollution and still guarantee crop productivity. The new materials should show better performance against weathering and/or better biodegradability once in the environment (EIP-AGRI Focus Group, 2021).

However, plastic pollution is not restricted to agricultural plastics but also includes littering and mismanaged plastic waste (MacLeod et al., 2021). Investing in the circular economy, increasing recycling strategies, raising public awareness, prohibiting disposable

plastics, and encouraging people to replace plastic with other materials could also help reduce plastic pollution (Borrelle et al., 2020; EIP-AGRI Focus Group, 2021; MacLeod et al., 2021).

These considerations reinforce that there is no single strategy that can fully address plastic pollution (Borrelle et al., 2020). Instead, solutions that consider context-specific combinations of reduction, recycling, the development of new materials, substitution, and mitigation strategies are required.

5.7 Limitations and future perspectives

The approach adopted in this thesis allows for a more comprehensive understanding of plastic degradation, accumulation, and transport. The evidence from the three chapters highlights that the use of plastic in agriculture, although necessary, poses an environmental problem. As demonstrated in Chapter 4, replacing conventional mulch films with soil-biodegradable mulch films is not very attractive without a full understanding of their environmental implications. Once in the environment, residues of both types of mulch film (conventional and biodegradable) can have irreversible effects on soil and surrounding ecosystems (Bandopadhyay et al., 2018; Kumar et al., 2020), as they are likely to persist. The smaller these residues become, the more difficult it is to recover them, and the more likely they are to interact with flora and fauna.

Laboratory experiments (Chapter 2) provide mechanistic insight into the photodegradation process and the influence of abrasion, but may not capture the full complexity of field conditions. Various factors can affect the photodegradation of agricultural plastics, and creating a laboratory test setting that mimics processes that occur under field conditions can be challenging. Additional parameters, such as temperature and humidity, should be included to provide a more realistic mechanism. The type of agricultural plastic should also be considered when conducting control experiments, as different plastics may have different formulations (e.g., different amounts of additives) to serve different purposes.

Field experiments (Chapter 3) demonstrated the influence of regional climates and management practices on plastic concentrations in the agricultural ecosystems. The concentration is influenced by multiple factors, including input sources, plastic characteristics, poor historical management practices documentation, regional climate, and so on. Since no long-term experimental site is available allowing analysis under controlled conditions, no standardised protocols for plastic detection are currently available, and several factors influencing plastic concentration, one way to increase comparability among studies is to be highly transparent in reporting data and methodologies. Even though soil matrices differ (e.g., organic matter content) and require different treatments to better detect plastic residues, as transparency in the data increases, data transformation and comparability can be performed and accounted for.

Heavy rainfall simulations (Chapter 4) showed the influence of freshly ploughed soil-biodegradable residues on surface runoff and their transport. However, the simulations had limitations regarding temporal scale, as they didn't account for long-term and seasonal effects, and regarding hydrological representativeness, as they didn't account for long-term soil-water processes. Rainfall simulations can be highly beneficial for enhancing our understanding of soil hydrological processes and the transport of residues; however, due to the limitations presented here, their accuracy and applicability can be restricted and need to be taken into perspective.

General conclusion and outlook 6

Agricultural plastics are essential for modern agriculture; however, they represent a persistent environmental pollution. This thesis puts into perspective the benefits of using agricultural plastics and the environmental pollution they cause. This thesis offers a valuable multi-scale perspective by combining laboratory and field experiment approaches to interlink the degradation, accumulation factors (i.e., climate and management), transport, and effects on soil processes of agricultural plastics. Moreover, it highlights the interactions among plastic properties, management practices, and environmental conditions. Understanding the connections between these factors is essential for predicting the behaviour, accumulation, and fate of agricultural plastic and, therefore, assessing environmental impacts.

It was demonstrated that agricultural plastic can present a rapid surface degradation, which can be accelerated by mechanical abrasion, contributing to microplastic formation (Chapter 2). However, different agricultural plastics behave differently, as they can differ in formulation to serve different purposes. Additionally, it becomes clear that factors other than degradation influence plastic accumulation (Chapter 3). Although the degradation mechanism presented (Chapter 2) can help explain the formation of plastic residues, other sources and factors, such as management practices (e.g., compost use) and environmental differences (e.g., regional climate), can also influence plastic residue concentrations in agricultural soil (Chapter 3). Moreover, during heavy rainfall simulations, soil-biodegradable residues increased surface runoff and influenced the transport of small residues (< 2 mm), potentially impacting surrounding ecosystems (Chapter 4). Although soil-biodegradable mulch films are a strong alternative to conventional mulch films, they raise environmental concerns that require further evaluation.

Although limitations were highlighted and future research is needed, our findings clearly advance our understanding of how agricultural plastics transition from indispensable agricultural tools to environmental pollutants, and the factors that drive their pollution and accumulation, showing how complex and dynamic these different factors are in influencing plastic pollution. The formation of plastic residues can happen due to different factors; however, their influence and impact can persist for long periods. If measures are not taken, agricultural plastic can threaten the environment, food productivity and safety.

Understanding the variables that influence the life cycle of agricultural plastics is important for understanding environmental effects, responsible use, and for developing better mitigation policies that balance increasing food productivity with its environmental risks. Nevertheless, without an effective and comprehensive monitoring system that accounts for regional environmental factors, regional management practices, and specific plastic properties, the environmental impact of both conventional and soil-biodegradable agricultural plastics cannot be fully understood and may be easily overlooked.

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Supplementary material

Chapter 2

Using the methodology described in the section “Analysing film properties”, the carbonyl index using the peak height values method (Figure S1) and the hydroxyl index using peak height values (Figure S2) and the area under the curve (Figure S3) methods were calculated.

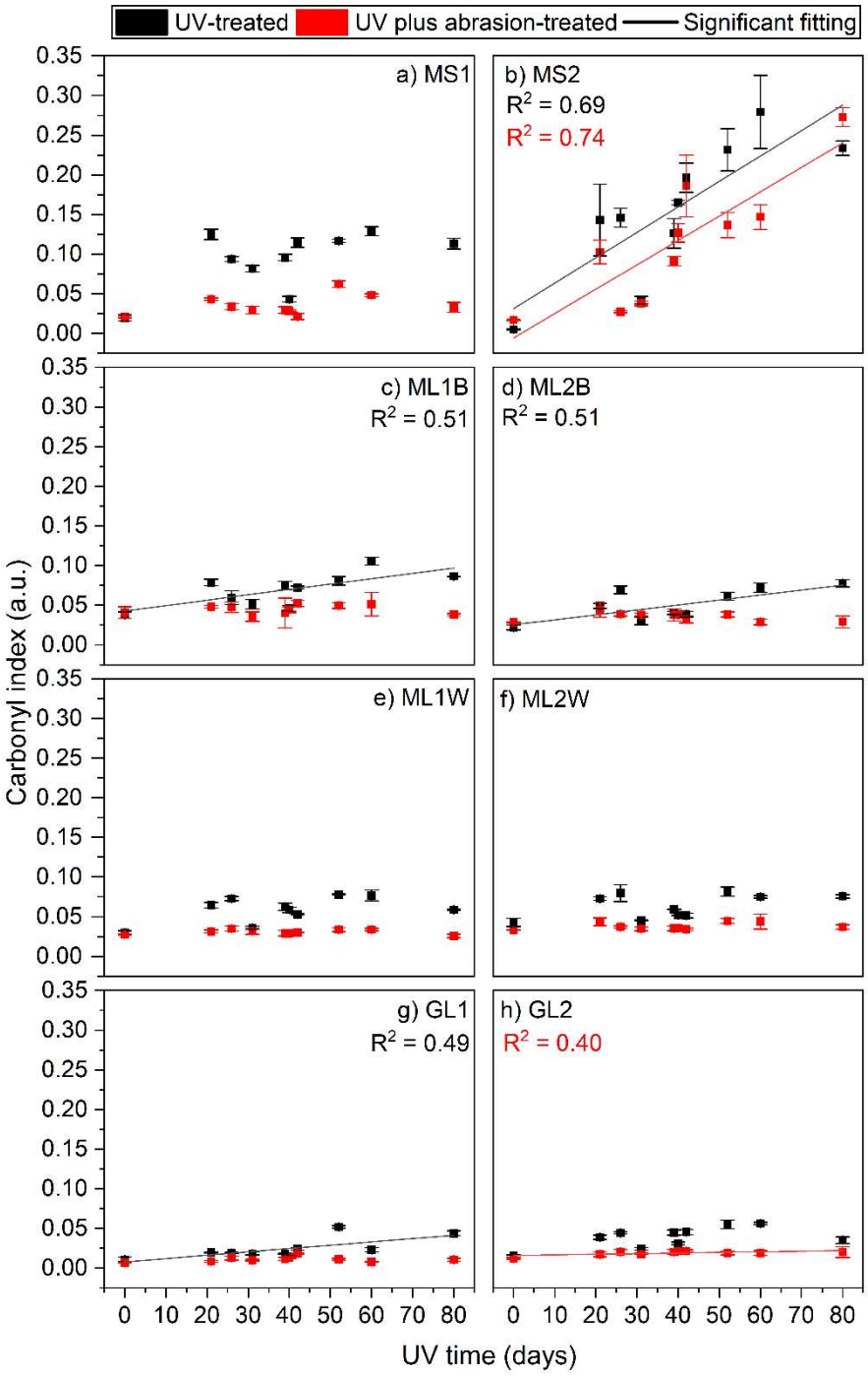

Figure S1: Carbonyl index of the different mulch film UV-treated and UV plus abrasion-treated samples using the peak height values method. For an overview of the abbreviation, please see Table 2.1.

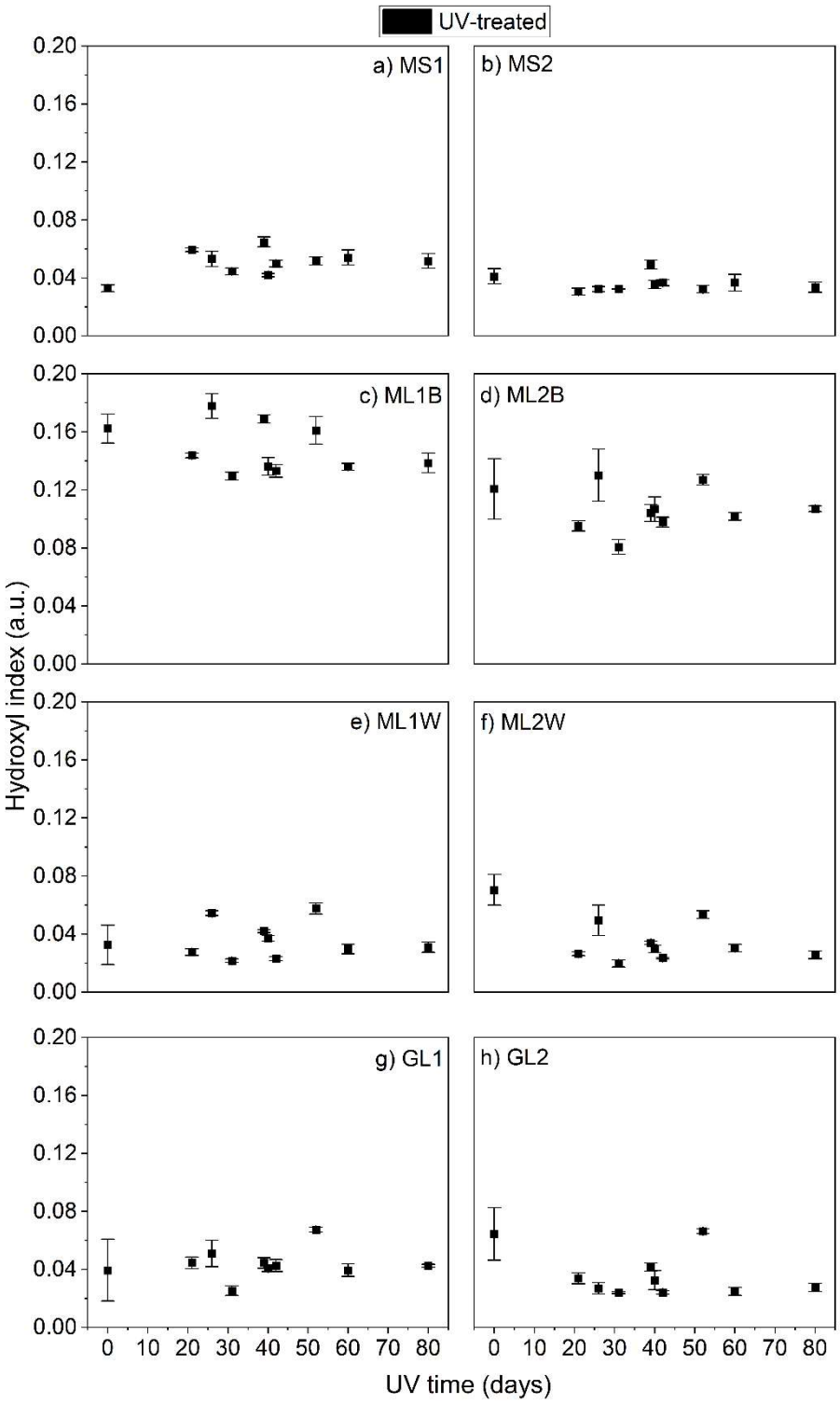

Figure S2: Hydroxyl index of the different mulch film UV-treated samples using the peak height values method. For an overview of the abbreviation, please see Table 2.1.

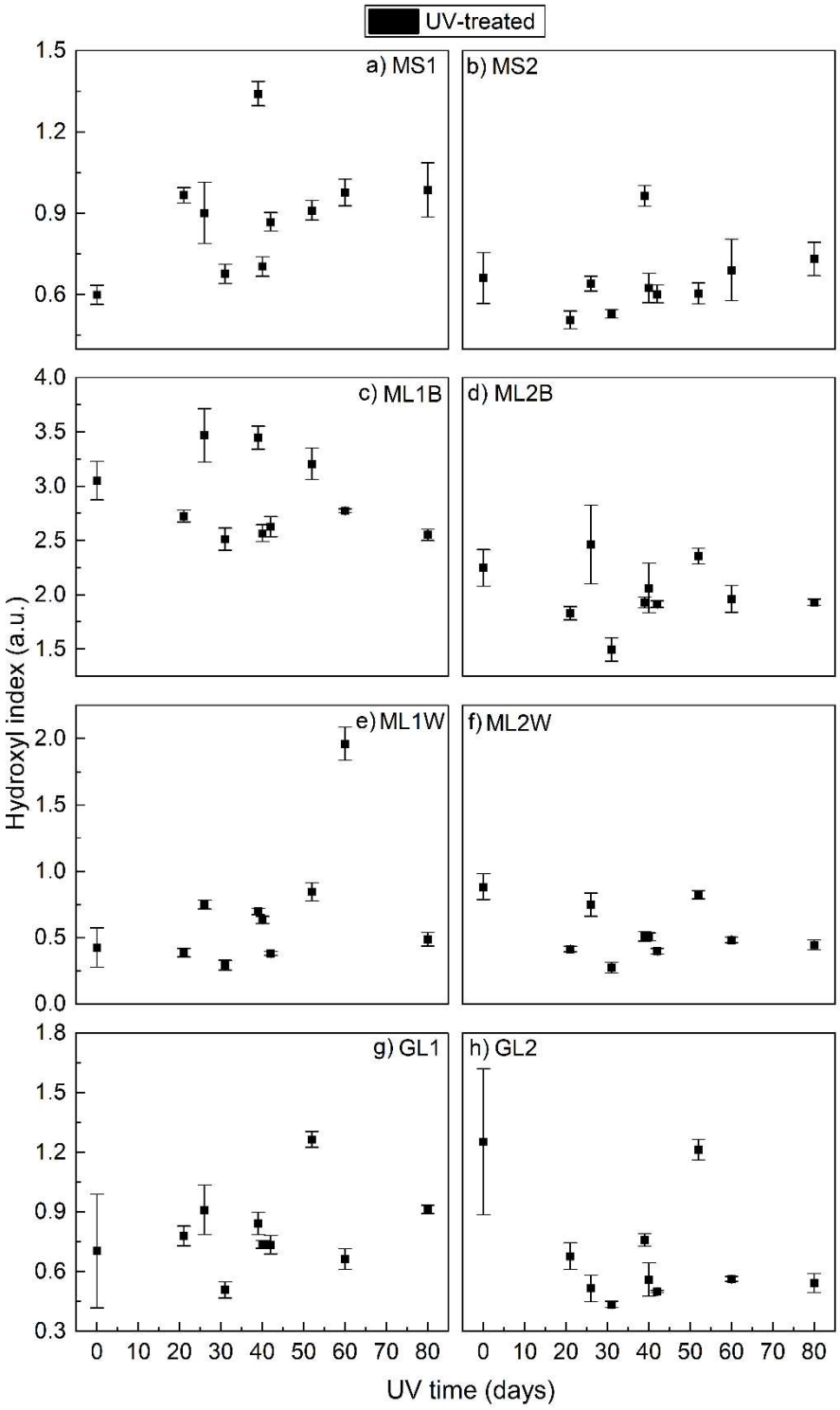

Figure S3: Hydroxyl index of the different mulch film UV-treated samples using the area under the curve method. For an overview of the abbreviation, please see Table 2.1.

Chapter 3

Table S1: Extended information on sampling area, crop, mulch removal time, and compost usage.

Field ID	Crop	Test site area (m x m)	Mulch removal time (year)	Compost	Note
S-ER1[a]	Raspberry	25x25	8-10	No info	Plastic was partially removed from the fields 20 years ago
S-ER2[a]	Raspberry	25x25	8-10	No info	Plastic was partially removed from the fields 20 years ago
S-ER3[a]	Raspberry	25x25	8-10	No info	Plastic was partially removed from the fields 20 years ago
S-LL1[b]	Beans	35x18	1	Yes	Soil changed in 2021
S-LL2[b]	Beans	35x18	1	Yes	Soil changed in 2021
G-MA1[b]	Mixed crops	25x25	-	200 t fresh matter in 2021	Use plastic accordingly to the crop
G-MA2[b]	Mixed crops, including asparagus	25x25	-	400 t fresh matter in 2021	Use plastic accordingly to the crop
G-MA3[b]	Mixed crops, including asparagus	25x25	-	No compost	-
G-NU1[b]	Mixed crops, including asparagus	12.5x25	-	30 t/ha (dry) every two years since 2000	Tunnels from 2012-2017 (30% of the field)
G-NU2[b]	Mixed crops, including asparagus	12.5x25	-	30 t/ha (dry) every two years since 2006	-
G-NU3[b]	Horseradish*	25x25	-	30 t once in 10 years	Black film tubes act as root barriers, restricting root spread. Crop every 4 years since 2008
G-NU4[b]	Horseradish*	25x50	-	30 t once in 10 years	Black film tubes act as root barriers, restricting root spread. One crop 2010
G-NU5[b]	Horseradish*	25x25	-	30 t once in 10 years	Black film tubes act as root barriers, restricting root spread. One crop 2017
G-NU6[b]	No crop	25x25	-	30 t once in 10 years	-
G-NU7[b]	No crop	25x25	-	30 t once in 10 years	-
G-NU8[b]	Horseradish*	25x25	-	30 t once in 10 years	Black film tubes act as root barriers, restricting root spread. Crop 2009 and 2021 (one section). Close to the highway
G-NU9[b]	Horseradish*	25x25	-	30 t once in 10 years	Black film tubes act as root barriers, restricting root spread. Crop 2007
G-NU10[b]	No crop	25x25	-	30 t once in 10 years	-
G-NU11[b]	No crop	25x25	-	No compost	-

- Information not available.

[a] Large-scale farm, more mechanised.

[b] Small-scale farms, less mechanised.

* Use of film tubes.

Chapter 4

^{1}H NMR spectra illustration.

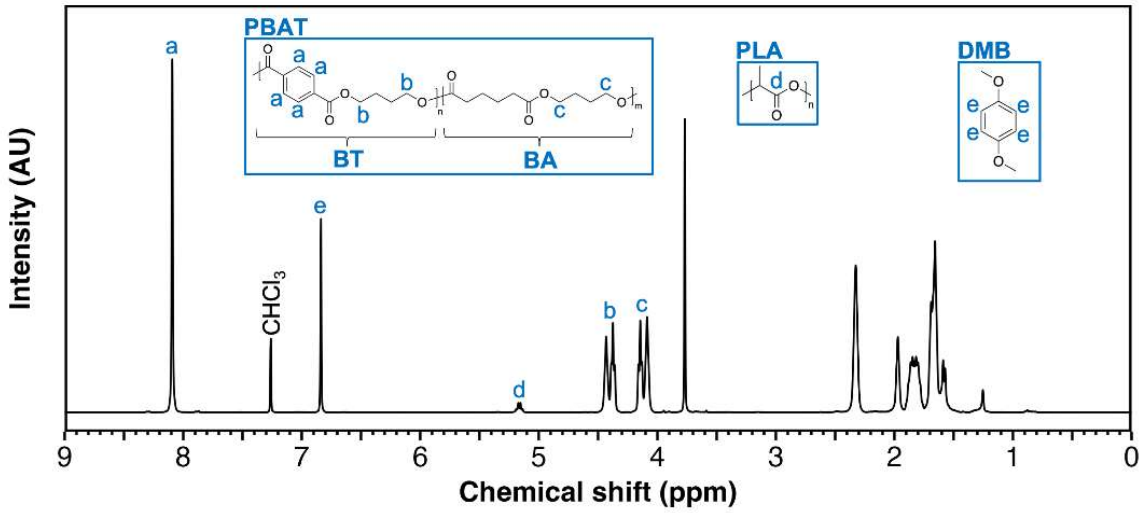

Figure S4: Exemplary proton nuclear magnetic resonance (^{1}H NMR) spectrum of biodegradable mulch film dissolved in deuterated chloroform ($CDCl_3$). The mulch film contains both polybutylene adipate-co-terephthalate (PBAT) and polylactic acid (PLA). The internal standard was dimethoxybenzene (DMB). Protons in the chemical structures and corresponding peaks in the spectra are annotated.

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